

Climate and plant traits drive a cross-continental imbalance in atmospheric Hg uptake

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This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

This manuscript examines trends in Hg₀ uptake by foliage across the globe. They begin by comparing concentrations of Hg in foliage to leaf age, and note a strong positive correlation. With this relationship, the authors model leaf age and biomass, then use this information to predict Hg₀ concentrations in foliage. Through structural equation models, they find that leaf properties are the most important variable to explaining leaf Hg concentrations and that regional Hg₀ concentrations are not good predictors. They report values for global forest Hg uptake that are much more constrained than previously reported ranges across an order of magnitude.

This study is incredibly robust and comprehensive. The compilation of leaf age and biomass data is an important contribution to the broader field of ecology, and the linking of these data to Hg₀ concentrations is done well. The approach is logical, and the results important and novel. I also want to commend the authors on an extremely well-written manuscript, with clear methods, results, and conclusions. Overall, I have few modifications to the manuscript.

There are three additional analyses that I think could improve the manuscript. First, the manuscript did not include any data near sources of Hg emissions. These locations likely have elevated local Hg₀ concentrations. It would be useful to determine whether local Hg₀ concentrations impact foliar Hg concentrations, in contrast to regional Hg₀ concentrations which they show to have no effect. This information is an important missing component of the manuscript, especially given that artisanal and small-scale gold mining occurs primarily in the tropics and emits Hg low to the ground, so is likely taken up by these trees. Secondly (though potentially out of the scope of this manuscript), it would be useful to link the litterfall Hg data to soil storage of Hg. Do the patterns observed in foliage and litterfall relate to soil Hg concentrations and pools? Third, it could be very interesting to model how predicted changes in foliar age, stomatal activity, and leaf biomass as a result of climate change might change the patterns and/or fluxes reported in the manuscript.

Line-by-line edits

Lines 41-42 – Incomplete sentence

Lines 46-47 – Please explain what is meant by “buffering”

Line 105 – What is meant by “direct anthropogenic effects”?

Lines 105-108 – It is unclear how this finding challenge the assumption that atmospheric Hg₀ concentrations dominate foliar Hg uptake.

Line 110 – This is the first figure referred to in the text, so should be figure 1. Also, it would be helpful to write out words on the figure, rather than using abbreviations

Lines 140-149 – Are the reported Hg₀ levels collected at the same location as the leaves? It is possible that leaf Hg is driven by local and not regional patterns in Hg₀. Co-located Hg₀ and leaf samples from these different regions would be more reliable than regional Hg₀ concentrations.

Lines 338-347 – Climate change is expected to change how CO₂ is taken up by leaves, as it changes patterns in stomatal activity. It would be useful to discuss and model how these hypothesized mechanisms would change Hg₀ uptake by foliage across the latitudinal gradient.

Lines 387-388 – It is unclear why foliar Hg concentrations near contaminated regions were not included. These would allow an assessment of local vs. regional sources of Hg₀ to foliage. Additional analyses incorporating these local sources of Hg₀ to determine the impact Hg₀ in foliage are necessary.

Lines 411 - Anthropogenic activities is not the best description of the traits of anthropogenic Hg emissions and atmospheric

Hg concentrations, as the latter is not necessarily due to anthropogenic activity.

SI Text – It would be useful to explain how leaf nitrogen and phosphorus contents are used in the leaf lifespan model.

References – At least one reference appears more than once (ie, the same reference has 2 numbers).

(Remarks on code availability)

I did not see the code or data to be able to evaluate it.

Reviewer #2

(Remarks to the Author)

The study presents a new global estimate of Hg uptake by forests, 1119 ± 329 Mg yr⁻¹, finding that tropical rainforests are the biggest vegetation sink for atmospheric Hg₀. The fact that tropical rainforests are major sinks for atmospheric Hg has been reported by other studies, including the Wang et al. (2016, ES&T) litterfall assessment by the same research group. The novel approach here is to calculate and account for leaf lifespan and leaf biomass, along with leaf Hg concentrations, in order to produce the estimates of forest uptake. It is not immediately clear to me how significant this methodological advance is compared to the Wang et al. (2016) approach, since the end results appear very similar (comparing Figure 5 in the 2016 paper with Figure 3 in the current study). Overall, I could see this study being a useful advance as an independent calculation of the global vegetation sink of Hg, but I think it is necessary to include further contextualisation of the results compared to existing studies and clarifications in the methodology. My major concerns would be:

1) The authors suggest that this study reduces the uncertainty in forest Hg uptake from 320–3138 Mg yr⁻¹ to 1119 ± 329 Mg yr⁻¹. To me, a wider uncertainty range still stands because other papers have used similar data-driven approaches with similar datasets, but due to different decisions in data treatment they report divergent estimates. It remains difficult to know which of these estimates is the “most accurate”, or if the variability in results is just due to a lack of measurement data and uncertainties in the modelling approach. I think further clarification between the data-driven approaches listed in Table 1 is required to provide further context for the current study’s results. I understand why the GEOS-Chem based estimates could differ from the current approach, but other data-driven studies like Chen et al. (2024) and Zhou and Obrist (2021) also estimate higher vegetation uptake (> 2000 Mg yr⁻¹) than in the current study, yet they use similarly comprehensive datasets of Hg in vegetation. Table 1 also lists two higher estimates from “this study” (1905 and 1604 Mg yr⁻¹) based on NPP that aren’t discussed at all in the text of this paper. Where do these come from and why are they discounted in the results and conclusions?

2) In a related comment, the authors mention an “artificial overestimation of tropical uptake” from past studies. However, it is not clear to me in the paper or the SI where this overestimation in tropical uptake derives from and why it is “artificial” and not just resulting from decisions in the methodologies. This allegation needs to be further expanded, since it may be the source of the discrepancies between the estimates in the current study and others in the literature.

3) One of the novelties of this study is the incorporation of leaf age into the calculation of forest uptake. This relies on the linear assumption of the foliar Hg accumulation rate over time. However, in Fig. 1c, it appears that none of this joint leaf age & concentration data is available from the Amazon or Africa, and for broadleaf trees all data is for leaves under 1 year of age. How do we know that accumulation effect doesn’t saturate at older ages for broadleaf trees?

4) The methods section in the main manuscript is too brief to properly understand the approach; the salient information should be transferred from the SI into the main manuscript. For example, Section S3 in the SI is repetitive with what is already in the methods section but it includes additional information that should be incorporated into the main manuscript.

5) Fig S25-36 - It says the model performance was evaluated on a test set, but in the methods it is stated that training is done through cross-validation. Was an independent evaluation dataset held separately as a test set after training? What percentage of the dataset was reserved and how was it selected? This should be clarified in the methods.

6) Figure 4 - This figure could be a bit confusing as the budget does not balance, since it only represents elemental Hg₀ in the Hg cycle. There is a deficit of 2000 Mg yr⁻¹, presumably coming from oxidation of Hg₀ to HgII. I also find confusing the manipulation of the Outridge et al. (2018) oceanic fluxes to produce this Hg₀ number, based on multiplying fluxes by a factor of 0.58 due to the isotopic source-tracing measurements in Jiskra et al. (2021). This assumption is inaccurate, since Outridge et al. (2018) refers to the net deposition flux (due to HgII dry and wet deposition to the ocean) and the net evasion flux (due only to Hg₀), with Jiskra et al. (2021) referring to 58% being the total of gross (i.e., one-way) deposition to the ocean. Thus, the budget in this figure leads to incorrect numbers resulting from mixing assumptions for net vs. gross fluxes and Hg₀ vs. HgII fluxes from different studies. I think it is misleading to modify the fluxes in this way, as these oceanic fluxes are not the focus of this work but presenting them in this Hg₀ budget could lead to these new estimates being compiled incorrectly by future studies.

Other specific comments:

L63 - Jiskra et al. (2018) did a back-of-the-envelope calculation for the period 1990–2010, not the last two decades. A more recent study (Feinberg et al., doi:10.1073/pnas.2401950121, 2024) that investigated the trend over 2005–2020 using the GEOS-Chem model suggested that foliar uptake is not a major driver of recent Hg₀ trends.

L82 - the GEOS-Chem parametrisation does not depend only on Hg₀ concentration and leaf area index, there are other meteorological and land surface parameters included in the parametrisation.

Fig 2 - With the Structural Equation Modelling presented in Figure 2, it doesn't make sense to me that in plot a) climate has a negative relationship with anthropogenic activities, but in plot b) it has a positive relationship. Why should this change if these are independent from the broadleaf vs. coniferous distinction between plot a and b?

L123- this must be a typo that deciduous broadleaves and evergreen needles have the exact same mean and standard deviation concentration ($31.1 \pm 18.8 \text{ ng g}^{-1}$)

L243 - judging from Fig. S13, the order of the numbers cited in the brackets should be switched. 30.4 refers to China and 44.4 refers to Brazil. For clarity, it would be good to specify this specifically in the brackets, since otherwise it is not apparent that no measurements are available from central Africa

-L275 - it should be emphasised somewhere that especially measurements in central Africa and tropical Southeast Asia are lacking in the database

-L280 - should be "inter-species" instead of "inter-specific"

-L292, 293 - it's unclear where the emissions estimate comes from or what this refers to (is it total Hg emissions? Only primary anthropogenic? Terrestrial re-emissions?). Also, from what I can find in the Gerson et al. 2022 paper, there are no direct field measurements of emissions in that study, I think the 45 t yr^{-1} estimate is based on a bottom-up inventory estimate (cited refs 54 and 55 in Gerson22 study). It should be specified here where these numbers come from.

-L294 - what is the basis of the regional emissions number being $181 \pm 54 \text{ t yr}^{-1}$? What year is this based on? What inventory is this based on?

-L320 - I can't find this 484 Mg yr^{-1} number in either the cited Outridge or Gerson papers. Where does it come from? Please specify the inventory used for calculating these numbers.

-The references for Gerson et al. (2022) are repeated (50 and 52) in the reference list

Table 1 - Yuan et al. 2024 didn't use GEOS-Chem, they used the Community Land Model-Hg (CLM5-Hg) within the Community Earth System Model (CESM).

-Section S3 - The clarity of the equations needs to be improved: 1) the authors denote i as age, but between t_1 and t_2 the age of the leaf will change. 2) Another issue in Section S3 is the "mass conservation principle" on Line 99: it is stated that difference between the foliar mass at time 1 and the foliar mass at time 2 is litterfall, but wouldn't another change in the mass be due to photosynthesis? 3) Eq. 9 has non-Latin characters.

-The SI could be reorganised for easier navigation. Figures are cited out of order in the text (e.g., Fig. S47 mentioned before Fig. S30 for the first time, many of these examples). The numbers given on the title page do not correspond to the actual numbers of Figures (7 vs. 47).

-Fig S2 - I wonder if the air Hg concentration vs. foliage Hg concentration would be compared for individual sites rather than lumped over continents whether a relationship between these variables would be more apparent. Sites in Tibet will be quite different in air Hg concentration compared to Eastern China, and sites in industrialised or contaminated areas of Europe will also show higher air concentrations than elsewhere. Also, please clarify whether the air Hg concentrations that were analysed come from measurements or a model simulation.

- The results in Fig S11 seem inconsistent to me with the approach used to calculate leaf age. The results show that the calculated foliar assimilation fluxes = 82% of litterfall flux (based on the slope of the fit). So that means that on average, litterfall has gained Hg from somewhere, e.g., post-depositional accumulation of atmospheric Hg. However, in other parts of the paper (e.g. SI L167-168), it is assumed that litterfall represents the cumulative Hg level at the end of the leaf lifespan (not including post-lifespan accumulation in Hg?)

-Fig. S40 and S41 - It looks like samples with higher Hg concentrations are generally underestimated by the model, and the residuals increase with higher predicted values. Could this not affect the global estimate of Hg uptake? This needs to be further clarified since the foliar Hg concentration is a critical variable for this study

-SI Table 3 - What is the source of the gridded atmospheric Hg concentration data? Is it a model simulation or based on observations? If so, which model and what are the conditions of this run? This needs to be described somewhere as it is relevant for the conclusion that air Hg concentrations do not play a notable role in the spatial pattern of foliar uptake.

(Remarks on code availability)
I did not see any code uploaded.

Reviewer #3

(Remarks to the Author)
This is a sophisticated modeling study which simulates global forest Hg uptake on a global scale, with a particular focus on

tropical forests. The authors state that unexpectedly, they find that (minimally industrialized) rainforests of South America and Africa exhibit the world's highest atmospheric Hg accumulation rates, and that these forests disproportionately sequester Hg released from industrialized regions. The study constrains global forest Hg uptake to $1119 \pm 329 \text{ Mg yr}^{-1}$, and the authors highlight a vulnerability of tropical forests to anthropogenic Hg inputs and climate-induced shifts in vegetation and terrestrial Hg reservoirs.

The study represents, in my view, the highly sophisticated and advanced modeling approach of atmospheric Hg uptake by vegetation, which globally is a dominant atmospheric Hg sink and is subsequently transferred to ecosystems via litterfall leading to ecosystem exposure to this pollutant. They achieve this by finding, and implementing, that "leaf traits", particularly accumulation rates of Hg in foliage combined with foliar lifespan, govern foliar Hg concentrations and hence uptake. This is not a new finding per se, and has been documented in previous studies that show that leaf accumulation rates, leaf age and functional plant types determine foliar Hg uptake and lead to inherent differences in Hg levels among different plant functional types (i.e., broadleaf evergreens, coniferous, and deciduous plants). What is unique here, however, is that the authors are able to refine these relationships using global databases and implement these into a global model of forest leaf lifespan. I am not an expert in carbon/vegetation modeling and estimates of forest/foliar lifespan, but I encourage the authors to ensure their modeled lifespan estimates are compared/discussed with previous results and publications on this topic as it is fundamental for their new modeling approach of Hg uptake. The approach is transformational and allows for an unprecedented spatial and temporal method to simulate forest Hg uptake across global ecosystems. On a minor note, I noted that the authors in several places, including in the abstract, use the term physiological controls; in my view, the correct terminology is "trait-based framework (or functional/morphological plant traits)", which the authors may want to use consistently throughout the manuscript as physiological data are not measured (only inferred).

Previous studies have already highlighted the role of tropical forests as globally important atmospheric Hg sinks and hotspots for Hg deposition, so this is not new. This study, however, now reconciles results of previous studies based on a different scaling approaches (e.g., using NPP data) and correct large overestimation in some of these studies. The new global estimates are now quite consistent with ground-based litterfall deposition measurements/estimates - this is an important result. I also believe the authors have done a good job explaining uncertainties, variability, and potential error sources, which I appreciate. Because their modeling approach critically relies on accurate parameterization of leaf age/lifespans, however, I would appreciate a clearer description on how leaf age was measured/obtained/implemented from experimental databases into their model. The study would also benefit from some discussion of other deposition processes and deposition pathways, such as wet or Hg(II) deposition processes (e.g., in lines 288-291, the authors mention canopy interception and foliar fixation rates, yet it is unclear what processes dominates so some references to underlying uptake mechanism would be helpful).

In lines 223-236, the authors discuss that annual uptake, litterfall, and net change in canopy (and woody pools) are near zero, so that globally and in tropical regions, these ecosystems are "stable, self-regulating interface that sustains the global balance between atmospheric Hg₀ absorption and terrestrial deposition. Was this result modelled, measured, or simulated? This is unclear to me. I don't disagree with this notion in principle, but the authors should be careful to not underestimate (at least locally and regionally) impacts of interannual variability, climate variability, forest successional stages, or extreme climate/weather and disturbance events.

My main concern with this study is with the interpretation and conclusion of results, particularly as they refer to fate of Hg and implication of this sink for tropical/global forests and global Hg cycling. The study exclusively focuses on the uptake and almost completely neglects discussing fate and emission/re-emission processes. The authors conclude that the tropics "disproportionately sequester Hg released from industrialized regions", "function as an unintended biogeochemical shield for the planet", that "absorbed Hg is largely stored in dynamic and climate-sensitive reservoirs, i.e. canopy, litter and organic-rich surface soils.", and that "legacy Hg has been redistributed via long-range transport and is now stored in remote tropical ecosystems". Yet, all these conclusions depend on understanding the fate and residence time of Hg in forests surface reservoirs and soils, after their original deposition via plant litterfall. The authors do not have data, nor do they discuss in detail, fate and turnover dynamics of Hg in forest soils, with the exception of mentioning one study from the the Peruvian Amazon which estimated that forest (floor?) emissions were in similar magnitudes as deposition. Do the authors suggest that this is the case across the tropics, and if so what is the basis is for this assumption? Most of the discussion points related to equilibrium between deposition and emissions, and associated fate of Hg, is really confusing to me, and needs to be revised (e.g., lines 306-314. "near steady-state exchange"; forests "act as self-regulating buffers rather than permanent sinks", "imply that terrestrial retention relies on intact canopy cover and sustained organic matter accumulation"). I cannot follow the underlying argumentation here. I recommend to consider an increasing data availability of soil Hg levels/pools, including from the tropics, in discussing fate and implication of high tropical atmospheric Hg deposition.

In summary, I find this an excellent and important study meritorious to be published in a highly visible journal such as Nature. The manuscript should be more specific about its limitations and conclusions, particularly as it refers to the importance of the findings for tropical forests health and long-term reservoirs/retention of global Hg pollution - this will critically depend on the stability of reservoir pools (surface organic layers and soils) which is not well considered/constrained in this study.

(Remarks on code availability)

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

My concerns have been adequately addressed, and I have no further comments.

(Remarks on code availability)

Reviewer #2

(Remarks to the Author)

The authors have carefully addressed the reviewer comments, and I generally agree with the revisions that have been made. I have two remaining concerns:

1) During the revision, several of the linear regression analyses were adjusted to force the line of best fit to go through the origin (e.g., Fig S15 and S21). This is slightly misleading, as the R² for these fits become artificially inflated when the zero-intercept condition is used. The R² for these lines are no longer representative of goodness of fit, because they are calculated with the sum of squares using the origin instead of the mean of the data. I also don't see a physical justification here, as opposed to other cases where a zero-intercept condition would be employed, e.g., that distance = velocity * time. In the case of these two figures, observed litterfall deposition is compared to modelled foliage assimilation, with no clear reason for a zero intercept. Any modelled quantity could have a systematic offset from observations, and thus a non-zero intercept is highly probable and should be considered in regression models.

2) In the new calculation of source-proximal areas, from Text S5 it looks like a dataset of all mining-impacted areas (Tang and Werner 2023) was used to extract areas over which to calculate the global emission-impacted flux. However, this dataset is a general dataset for mining and thus not specific to ASGM locations. I think a further clarification is needed of which emissions source impacts are treated in this calculation. Is it just meant to be ASGM, in which case a specific ASGM geographical distribution should be considered; or are other contaminated forest sites considered, in which case why is this general mining-impacted areas map an appropriate dataset to capture Hg contamination?

(Remarks on code availability)

I have seen the code upload and it looks thorough, although I have not reviewed this in depth.

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RESPONSE TO REVIEWERS' COMMENTS

We thank the reviewers for their careful evaluation of the manuscript and for their constructive comments. The manuscript has been revised accordingly, and all comments are addressed below. Reviewer comments are shown in **BLACK**, followed by our responses in **BLUE**. Revisions in the manuscript are indicated with section and line numbers.

The revisions mainly include clarification of the modelling framework, additional explanation of methodological assumptions, and improvements to the presentation of results and uncertainties. In response, we have substantially revised the manuscript to clarify the modeling framework, strengthen model validation, and refine the interpretation of tropical forests as major sinks in the global Hg cycle. Major revisions include:

- (1) additional analysis of source-proximal regions;
- (2) expanded validation using litterfall and throughfall datasets;
- (3) clarification of the steady-state assumption between canopy uptake and litterfall deposition;
- (4) improved discussion of Hg fate in tropical soils and potential remobilization under disturbance; and
- (5) expanded methodological description of the leaf lifespan modeling framework.

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

This manuscript examines trends in Hg⁰ uptake by foliage across the globe. They begin by comparing concentrations of Hg in foliage to leaf age, and note a strong positive correlation. With this relationship, the authors model leaf age and biomass, then use this information to predict Hg⁰ concentrations in foliage. Through structural equation models, they find that leaf properties are the most important variable to explaining leaf Hg concentrations and that regional Hg⁰ concentrations are not good predictors. They report values for global forest Hg uptake that are much more constrained than previously reported ranges across an order of magnitude.

This study is incredibly robust and comprehensive. The compilation of leaf age and biomass data is an important contribution to the broader field of ecology, and the linking of these data to Hg⁰ concentrations is done well. The approach is logical, and the results important and novel. I also want to commend the authors on an extremely well-written manuscript, with clear methods, results, and conclusions. Overall, I have few modifications to the manuscript.

Response:

We thank the reviewer for the constructive comments, which helped improve the manuscript.

There are three additional analyses that I think could improve the manuscript. First, the manuscript did not include any data near sources of Hg emissions. These locations likely have elevated local Hg⁰ concentrations. It would be useful to determine whether local Hg⁰ concentrations impact foliar Hg concentrations, in contrast to regional Hg⁰ concentrations which they show to have no effect. This information is an important missing component of the manuscript, especially given that artisanal and small-scale gold mining occurs primarily in the tropics and emits Hg low to the ground, so is likely taken up by these trees.

Response:

We thank the reviewer for this insightful suggestion. To evaluate the influence of elevated local Hg⁰ concentrations, we expanded the dataset to include foliar Hg measurements from regions located near major Hg emission sources, including areas influenced by artisanal and small-scale gold mining (ASGM) in tropical regions. Using these additional data, we developed a modeling framework to estimate foliar Hg⁰ uptake in source-proximal forests.

The analysis indicates that foliar Hg⁰ uptake in emission-influenced regions amounts to $86 \pm 42 \text{ Mg yr}^{-1}$. The highest uptake occurs in South America, Southeast Asia, and the United States ($15\text{--}25 \text{ Mg yr}^{-1}$), while other regions show lower uptake ($4\text{--}7 \text{ Mg yr}^{-1}$). After incorporating this additional component, the updated global forest foliar Hg⁰ sink is estimated at $1188 \pm 438 \text{ Mg yr}^{-1}$.

These results show that although local emission sources can enhance foliar Hg concentrations near source areas, the dominant control on global-scale foliar Hg uptake remains vegetation traits and climate, consistent with the main conclusions of this study.

Revision in manuscript (Section 2.3, Lines 215–222):

These additional analyses and datasets have been incorporated into the revised manuscript (Text S5 and Fig. S10). Regional statistics of foliar Hg uptake in source-affected areas are presented in Fig. S11.

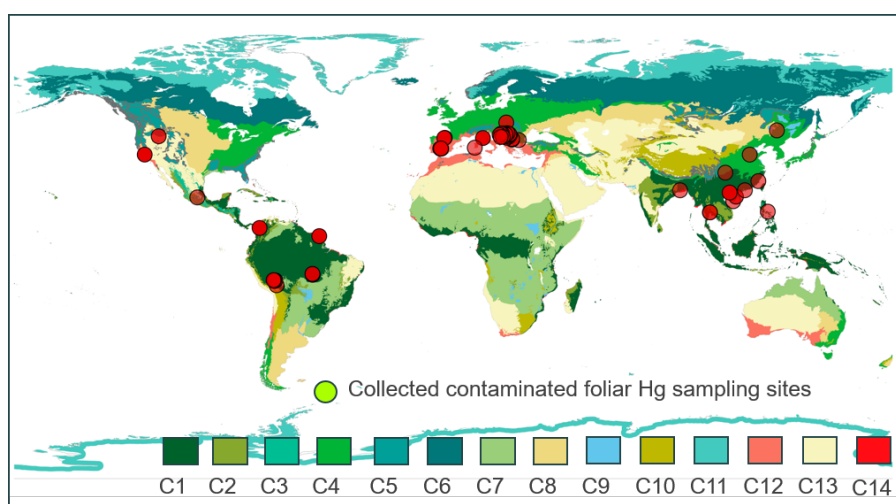


Figure S10. Spatial distribution of foliar Hg samples collected from contaminated sites across the 14 WWF (World Wildlife Fund) terrestrial ecoregions.

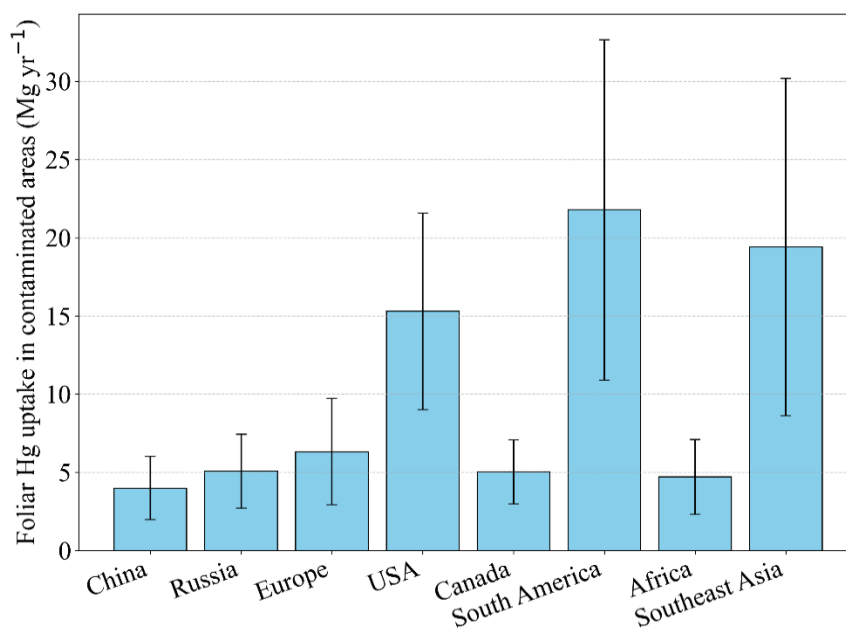


Figure S11. Regional statistics of foliar Hg^0 uptake in source-affected (contaminated) areas.

Secondly (though potentially out of the scope of this manuscript), it would be useful to link the litterfall Hg data to soil storage of Hg. Do the patterns observed in foliage and litterfall relate to soil Hg concentrations and pools?

Response:

We thank the reviewer for this insightful suggestion. To explore the link between canopy Hg inputs and soil Hg storage, we conducted a preliminary comparison between our modeled foliar Hg⁰ uptake and published datasets of surface soil Hg pools. The analysis reveals a clear spatial correspondence: both global foliar Hg⁰ uptake and surface soil Hg storage (0-5 cm depth) exhibit similar latitudinal gradients, decreasing from the equator toward high-latitude boreal regions (Fig. S23). This pattern suggests that foliar Hg uptake, followed by transfer to soils through litterfall, is a major pathway governing Hg accumulation in surface organic soil horizons.

These results support the conceptual framework that forests act as an important conduit transferring atmospheric Hg to terrestrial ecosystems. They also indicate that changes in vegetation structure or productivity—particularly in tropical forests—may influence both current Hg uptake and the stability of Hg stored in surface soils.

Revision in manuscript (Section 3, Lines 390–397):

A new paragraph and corresponding analysis have been added to discuss the relationship between foliar Hg uptake and surface soil Hg storage (Fig. S23).

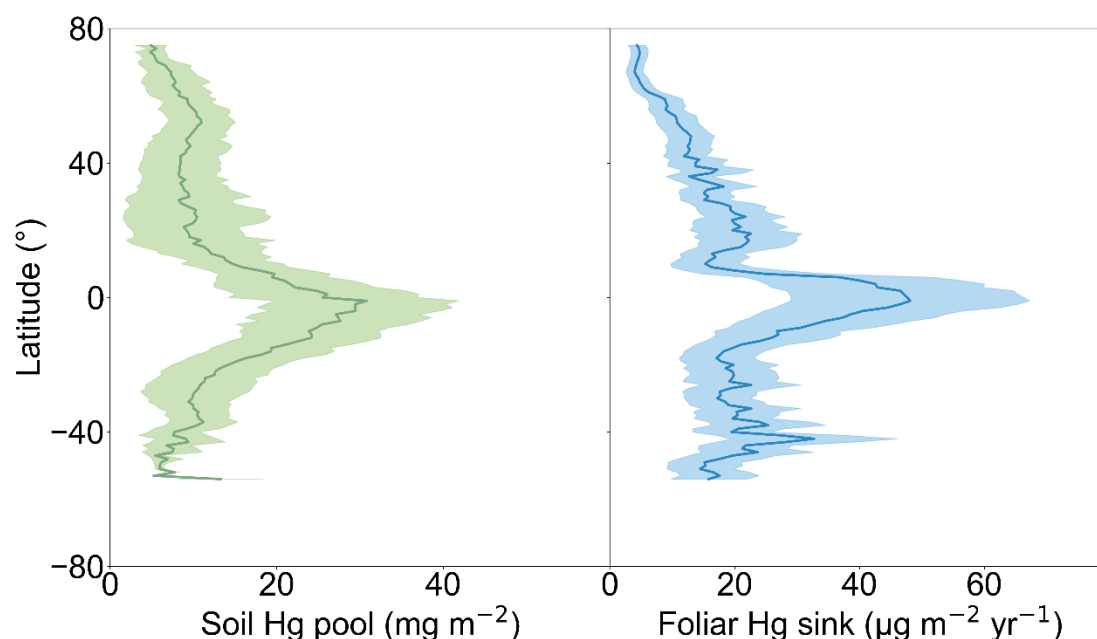


Figure S23. Latitudinal distribution of soil Hg pools⁴⁰ and modelled foliar Hg⁰ uptake. The aligned latitudinal patterns suggest that foliar uptake followed by litterfall transfer governs surface soil Hg accumulation.

Third, it could be very interesting to model how predicted changes in foliar age, stomatal activity, and leaf biomass as a result of climate change might change the

patterns and/or fluxes reported in the manuscript.

Response:

We thank the reviewer for this excellent and forward-looking suggestion. We agree that assessing how climate change may influence foliar Hg uptake through changes in vegetation traits and climatic drivers is an important direction for future research. To address this point, we have incorporated a simplified scenario analysis into the revised manuscript and Supplementary Information (Text S12).

Simulating the future forest Hg⁰ sink is inherently complex because it depends on multiple interacting factors, including atmospheric Hg concentrations, anthropogenic emissions, climatic conditions, and vegetation traits. As an initial exploration, we conducted a simplified scenario analysis using downscaled temperature and precipitation projections from the IPSL climate model under the SSP1–2.6 scenario to evaluate the isolated effects of projected climate change on the global forest Hg⁰ sink for the period 2081–2100.

The results show that changes in precipitation alone produce negligible effects on the global forest Hg⁰ sink (<0.2%, Fig. S25), with future-to-present ratios of leaf lifespan and biomass remaining close to unity (0.98–1.02). When only temperature was modified, the overall global response remained small (~1%, Fig. S26), but with distinct regional variations. In high-latitude boreal regions (60–70°N), projected warming leads to a slight reduction of the forest Hg sink (~3 Mg yr⁻¹), primarily due to shortened leaf lifespan and reduced Hg accumulation duration. In contrast, temperate regions of the Northern Hemisphere show an increase of approximately 15 Mg yr⁻¹, where warming-enhanced foliar uptake likely outweighs reductions associated with shorter leaf lifespan or biomass. In low-latitude forests, projected temperature changes are small (<10%), resulting in minimal changes in foliar traits and therefore limited effects on the forest Hg⁰ sink.

Overall, these simplified scenario analyses highlight the regionally variable responses of forest Hg cycling to climate change. While a full assessment would require coupled simulations integrating vegetation dynamics, atmospheric Hg concentrations, and land-use change, these results indicate that climate-driven vegetation responses may either reinforce or offset emission-reduction benefits, complicating future evaluations of the Minamata Convention.

Revision in manuscript (Section 3, Lines 443–464):

A new scenario analysis examining the potential effects of future climate change on the global forest Hg⁰ sink has been added to the revised manuscript and Supplementary Information (Text S12; Figs. S25–S26).

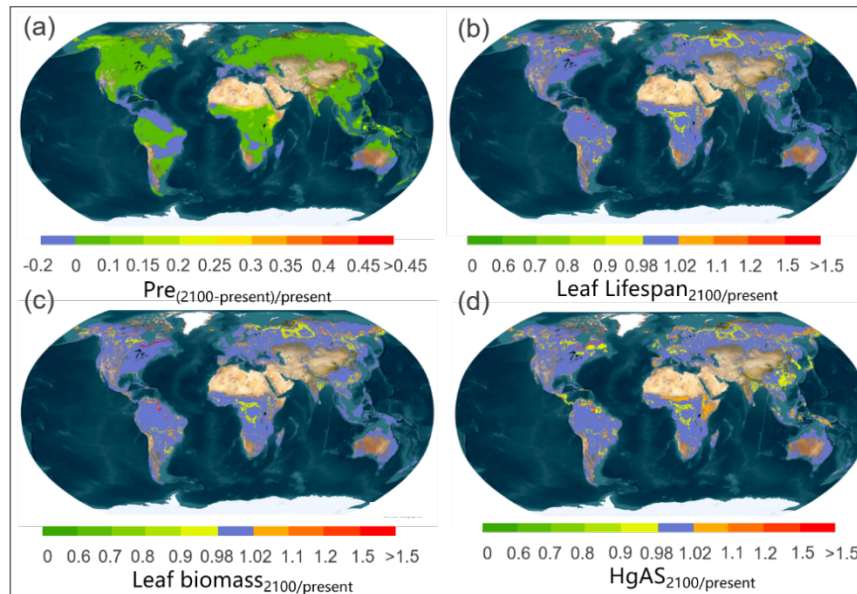


Figure S25. Projected changes in forest Hg^0 uptake under SSP1-2.6 driven by precipitation only (2081–2100): **(a)** relative change in precipitation ((future – present)/present); **(b)** leaf lifespan; **(c)** leaf biomass; **(d)** foliar Hg^0 uptake. Minimal changes indicate that precipitation alone exerts limited control on future forest Hg^0 uptake.

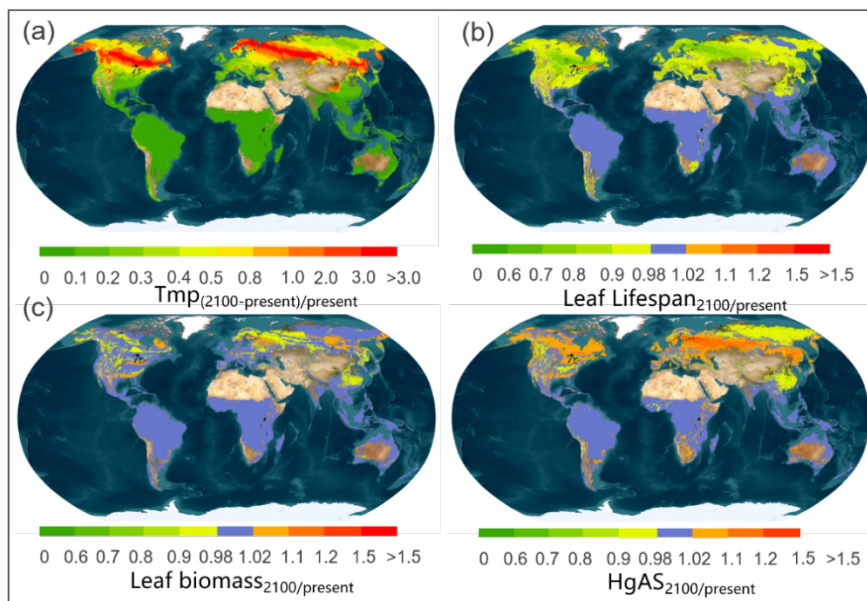


Figure S26. Projected changes in forest Hg^0 uptake under SSP1-2.6 driven by temperature only (2081–2100): **(a)** relative change in temperature ((future – present)/present); **(b)** leaf lifespan; **(c)** leaf biomass; **(d)** foliar Hg^0 uptake. Temperature effects are modest overall but drive contrasting regional responses in forest Hg^0 uptake.

Line-by-line edits

Lines 41-42 – Incomplete sentence

Response:

This Incomplete sentence has been revised

Lines 46-47 – Please explain what is meant by “buffering”

Response:

Here, we mean mitigating the spread of anthropogenic contaminants

Line 105 – What is meant by “direct anthropogenic effects”?

Response:

“Direct anthropogenic effects” refers to the impact of contemporary anthropogenic Hg emissions that elevate atmospheric Hg concentrations, thereby affecting foliar Hg concentrations. This has been further clarified in the revised manuscript.

Lines 105-108 – It is unclear how this finding challenge the assumption that atmospheric Hg⁰ concentrations dominate foliar Hg uptake.

Response:

We thank the reviewer for pointing out this ambiguity. The text has been revised to clarify how our findings refine the conventional view that atmospheric Hg⁰ concentrations dominate foliar Hg uptake. Our results show that foliar Hg content is not determined by atmospheric Hg⁰ concentrations alone, but is also strongly influenced by Hg accumulation rates and leaf lifespan—traits largely regulated by climate and vegetation physiological characteristics. This clarification emphasizes that plant functional traits, mediated by climatic conditions, play a dominant role in controlling foliar Hg accumulation across global forests.

Revision in manuscript (Section 2.1, Lines 116–126):

The text has been revised to clarify the role of vegetation traits and climate in regulating foliar Hg accumulation.

Line 110 – This is the first figure referred to in the text, so should be figure 1. Also, it would be helpful to write out words on the figure, rather than using abbreviations

Response:

It has been revised as suggested.

Lines 140-149 – Are the reported Hg⁰ levels collected at the same location as the leaves? It is possible that leaf Hg is driven by local and not regional patterns in Hg⁰. Co-located Hg⁰ and leaf samples from these different regions would be more reliable than regional. Hg⁰ concentrations.

Response:

We thank the reviewer for this important methodological clarification. We agree that co-located measurements of atmospheric Hg⁰ and foliar Hg provide the most reliable basis for assessing their relationship. To address this point, we analyzed paired

observations of atmospheric Hg^0 concentrations and foliar Hg collected at the same sites.

The results reveal a clear threshold behavior: when atmospheric Hg^0 concentrations exceed 5 ng m^{-3} , they exert a significant control on foliar Hg content ($p < 0.01$), whereas at concentrations below 5 ng m^{-3} the relationship is not significant ($p > 0.05$). Because atmospheric Hg^0 concentrations in most global forest regions are typically below this threshold, regional-scale atmospheric Hg^0 levels do not significantly influence foliar Hg concentrations.

Revision in manuscript (Section 2.1, Lines 116–123):

The text has been revised to clarify the analysis based on paired atmospheric Hg^0 and foliar Hg observations and the resulting threshold relationship (Fig. S1; Figs. S2–S3).

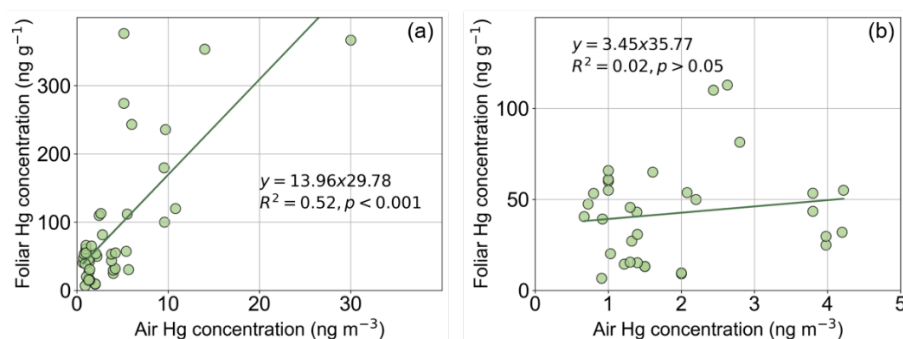


Figure S1. Relationship between atmospheric Hg concentration and foliar Hg concentration derived from co-located field measurements. (a) Relationship across the full atmospheric Hg concentration range. (b) Relationship restricted to background atmospheric Hg conditions ($0\text{--}5 \text{ ng m}^{-3}$). These results indicate that atmospheric Hg^0 constrains foliar Hg only above a threshold, while below it other controls dominate.

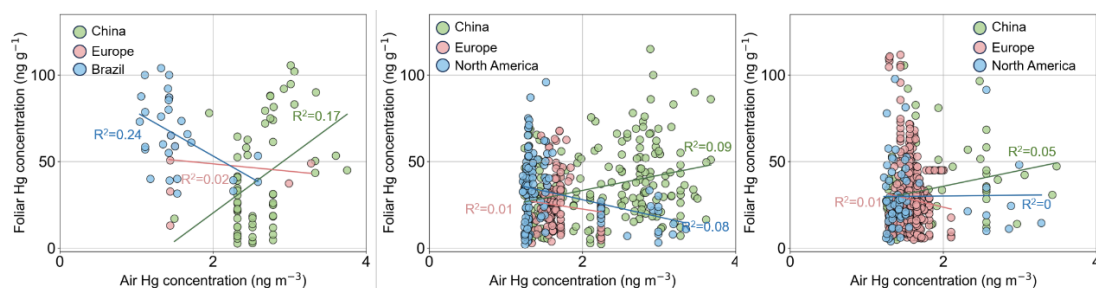


Figure S2. Relationships between foliar Hg concentrations and modeled atmospheric Hg^0 concentrations across different regions and forest types. Panel (a) shows evergreen broadleaf forests, panel (b) shows deciduous broadleaf forests and panel (c) shows evergreen needleleaf forests. These relationships show weak or absent correlations under background conditions, indicating limited regional control by atmospheric Hg^0 .

Lines 338-347 – Climate change is expected to change how CO_2 is taken up by leaves, as it changes patterns in stomatal activity. It would be useful to discuss and model how these hypothesized mechanisms would change Hg^0 uptake by foliage across the

latitudinal gradient.

Response:

We thank the reviewer for this insightful comment regarding the potential influence of rising atmospheric CO₂ on foliar Hg⁰ uptake through changes in stomatal conductance. We agree that this mechanism is important and should be discussed. However, quantitatively incorporating CO₂ effects into the current modeling framework remains challenging due to limited observational constraints linking CO₂, stomatal behavior, and foliar Hg dynamics at ecosystem scales.

Revision in manuscript (Section 3, Lines 453–460):

The manuscript and Supplementary Information (Text S12) have been revised to discuss the potential influence of rising CO₂ on forest Hg⁰ uptake. The revised text states that elevated CO₂ may induce partial stomatal closure and reduce gas exchange, potentially suppressing foliar Hg⁰ uptake. However, the magnitude of this effect remains uncertain because existing evidence largely derives from short-term experiments on individual plants, and it is unclear how these responses scale to mature forest canopies. Moreover, stomatal conductance is co-regulated by multiple environmental drivers, including water availability, temperature, and light. Consequently, future changes in temperature and precipitation may partially offset CO₂-induced stomatal closure. Given these uncertainties and the current lack of coordinated observations linking CO₂, stomatal conductance, and foliar Hg dynamics across global forests, CO₂ effects were not quantitatively incorporated into the present modeling framework.

Lines 387-388 – It is unclear why foliar Hg concentrations near contaminated regions were not included. These would allow an assessment of local vs. regional sources of Hg⁰ to foliage. Additional analyses incorporating these local sources of Hg⁰ to determine the impact Hg⁰ in foliage are necessary.

Response:

We thank the reviewer for this valuable suggestion. In response, we have incorporated additional analyses of foliar Hg concentrations in source-proximal regions to evaluate the influence of local Hg⁰ emissions on foliar Hg accumulation. The corresponding results and figures have been added to the revised manuscript and Supplementary Information. These additions allow a clearer assessment of the relative roles of local emission sources and regional atmospheric Hg⁰ concentrations in controlling foliar Hg levels.

Lines 411 - Anthropogenic activities is not the best description of the traits of anthropogenic Hg emissions and atmospheric Hg concentrations, as the latter is not necessarily due to anthropogenic activity.

Response:

Thank you for this helpful comment. We agree that the term “anthropogenic activities” was imprecise, as atmospheric Hg concentrations reflect both anthropogenic emissions and background processes. In the revised manuscript, this component has been clarified and is now described as “direct anthropogenic effects”. In addition, the corresponding description has been moved from the main text to the Supplementary Information (Text S1), where the SEM framework is presented in detail. The revised wording more accurately distinguishes between direct anthropogenic emissions and atmospheric Hg concentrations as a combined signal of multiple sources.

SI Text – It would be useful to explain how leaf nitrogen and phosphorus contents are used in the leaf lifespan model.

Response:

We thank the reviewer for this helpful suggestion. We agree that clarifying the role of leaf nitrogen (N) and phosphorus (P) in the leaf lifespan model improves the transparency and mechanistic basis of our approach. To address this point, we expanded the explanation in the Supplementary Information.

Leaf N and P are key plant functional traits closely linked to photosynthetic capacity and metabolic rates, and they are widely recognized predictors of leaf lifespan. Previous studies have demonstrated a fundamental trade-off in plant resource economics: leaves with higher N and P contents generally exhibit higher photosynthetic rates and faster tissue turnover, resulting in shorter leaf lifespans, whereas leaves with lower N and P contents tend to have slower metabolic rates and longer longevity. Incorporating foliar N and P into our modeling framework therefore allows established plant physiological relationships to constrain the spatial patterns of leaf lifespan, which in turn improves estimates of foliar Hg accumulation time and Hg⁰ uptake.

Revision in manuscript (Supplementary Information, Text S2, Lines 67–76):

Additional explanation has been added describing how foliar N and P are used as predictors of leaf lifespan within the modeling framework.

References – At least one reference appears more than once (ie, the same reference has 2 numbers).

Response:

We thank the reviewer for pointing this out. The reference list has been carefully checked and corrected to remove duplicate entries and ensure that each reference appears only once in the manuscript.

Reviewer #1 (Remarks on code availability):

I did not see the code or data to be able to evaluate it.

Response: We thank the reviewer for this comment. The code used in this study has now been uploaded and is made available for evaluation and reproducibility.

Reviewer #2 (Remarks to the Author):

The study presents a new global estimate of Hg uptake by forests, 1119 ± 329 Mg yr⁻¹, finding that tropical rainforests are the biggest vegetation sink for atmospheric Hg⁰. The fact that tropical rainforests are major sinks for atmospheric Hg has been reported by other studies, including the Wang et al. (2016, ES&T) litterfall assessment by the same research group. The novel approach here is to calculate and account for leaf lifespan and leaf biomass, along with leaf Hg concentrations, in order to produce the estimates of forest uptake. It is not immediately clear to me how significant this methodological advance is compared to the Wang et al. (2016) approach, since the end results appear very similar (comparing Figure 5 in the 2016 paper with Figure 3 in the current study). Overall, I could see this study being a useful advance as an independent calculation of the global vegetation sink of Hg, but I think it is necessary to include further contextualisation of the results compared to existing studies and clarifications in the methodology.

Response:

We thank the reviewer for this thoughtful comment. We agree that clarifying the methodological advances of our study relative to previous approaches, particularly Wang et al. (2016), is essential for demonstrating the novelty and significance of this work. To address this point, we substantially revised both the manuscript and the Supplementary Information (Text S10) to clarify the conceptual and methodological differences between the two approaches.

First, the two studies quantify different components of the forest Hg cycle. Wang et al. (2016) estimated Hg deposition through litterfall, which indirectly reflects foliar Hg⁰ uptake under the assumption that annual canopy uptake is balanced by litterfall export. In contrast, our study directly resolves foliar Hg⁰ uptake by integrating foliar Hg accumulation rates with independently constrained leaf biomass and leaf lifespan.

Second, the methodological frameworks differ in their ability to identify mechanistic drivers. The litterfall-based synthesis of Wang et al. (2016) extrapolated deposition within biome classifications, whereas our study employs a trait-informed machine-learning cascade framework linking climate, anthropogenic activity, and plant functional traits to foliar Hg accumulation and uptake. This framework allows us to identify the environmental and physiological controls underlying the spatial heterogeneity of the forest Hg sink.

Third, our framework substantially reduces uncertainty in global estimates. The litterfall-based estimate of 1180 ± 710 Mg yr⁻¹ reported by Wang et al. (2016) has a relative uncertainty of ~60%, whereas our estimate of 1188 ± 438 Mg yr⁻¹ reduces this uncertainty to ~30%. This improvement results primarily from explicitly constraining leaf biomass and leaf lifespan, which regulate the residence time and accumulation of Hg in foliage.

Revision in manuscript:

- **Introduction (Lines 80–85):**
The introduction has been revised to clarify the limitations of previous litterfall-based approaches for estimating foliar Hg uptake.
- **Results / Section 2.3 (Lines 251–266):**
A new paragraph has been added explaining the conceptual differences between litterfall-derived Hg deposition estimates and our trait-based canopy uptake framework.
- **Results / Section 2.3 (Lines 201–205):**
The description of the modeling framework has been revised to emphasize that the mapping of global foliar Hg⁰ uptake explicitly integrates spatially variable leaf biomass and leaf lifespan.
- **Supplementary Information (Text S10):**
Additional discussion has been added to clarify the methodological advances of the present framework relative to Wang et al. (2016).

My major concerns would be:

1) The authors suggest that this study reduces the uncertainty in forest Hg uptake from 320–3138 Mg yr⁻¹ to 1119 ± 329 Mg yr⁻¹. To me, a wider uncertainty range still stands because other papers have used similar data-driven approaches with similar datasets, but due to different decisions in data treatment they report divergent estimates. It remains difficult to know which of these estimates is the “most accurate”, or if the variability in results is just due to a lack of measurement data and uncertainties in the modelling approach. I think further clarification between the data-driven approaches listed in Table 1 is required to provide further context for the current study’s results. I understand why the GEOS-Chem based estimates could differ from the current approach, but other data-driven studies like Chen et al. (2024) and Zhou and Obrist (2021) also estimate higher vegetation uptake (> 2000 Mg yr⁻¹) than in the current study, yet they use similarly comprehensive datasets of Hg in vegetation. Table 1 also lists two higher estimates from “this study” (1905 and 1604 Mg yr⁻¹) based on NPP that aren’t discussed at all in the text of this paper. Where do these come from and why are they discounted in the results and conclusions?

Response:

We acknowledge that several data-driven approaches using similar datasets have produced divergent estimates of global vegetation Hg uptake (Table 1). This variability largely reflects methodological differences rather than fundamental disagreement in the underlying observations. To clarify this issue, we revised the manuscript to explicitly explain the methodological differences among existing approaches and to justify the framework adopted in this study.

First, mechanistic atmospheric models such as GEOS-Chem and CESM2-CLM5 report a wide range of global vegetation Hg uptake estimates (320–3138 Mg yr⁻¹; Table 1). These models primarily parameterize vegetation uptake as a function of atmospheric Hg concentrations and meteorological variables, while representing vegetation physiological processes only indirectly. As a result, simulated spatial patterns tend to follow anthropogenic emission gradients, with high uptake predicted in emission hotspots such as eastern China (56–70 µg m⁻² yr⁻¹) and comparatively low uptake in tropical forests (21–42 µg m⁻² yr⁻¹). These patterns are inconsistent with observational evidence from litterfall and foliar measurements showing that tropical forests exhibit the highest Hg uptake globally despite relatively low atmospheric Hg concentrations. In contrast, our estimates (15–30 µg m⁻² yr⁻¹ for eastern China and 40–70 µg m⁻² yr⁻¹ for the Amazon and central Africa) closely match observed litterfall deposition fluxes (China: 30.4 ± 16.9 µg m⁻² yr⁻¹; Brazil: 44.4 ± 5.7 µg m⁻² yr⁻¹; Fig. S17).

Second, statistical approaches that estimate foliar Hg uptake by multiplying foliar Hg concentrations by leaf net primary production (NPP) are subject to two important biases. Multi-year cumulative foliar Hg concentrations represent Hg accumulated over the full leaf lifespan; multiplying these values by annual NPP therefore treats multi-year accumulation as if it occurred within a single year, leading to systematic overestimation. In addition, leaf NPP is typically derived by multiplying total aboveground NPP by fixed leaf allocation coefficients, although the fraction of biomass allocated to foliage varies substantially across forest types and climatic gradients. To clarify this issue, we explicitly state in the revised manuscript that the higher NPP-based estimates shown in Table 1 are presented only as methodological comparisons illustrating the magnitude of overestimation:

“For transparency, the higher NPP-based estimates listed in Table 1 for this study are presented only as methodological comparisons, illustrating the degree of overestimation that occurs when cumulative foliar Hg concentrations are multiplied by annual productivity.”

The higher estimate listed in Table 1 for this study (1654 Mg yr⁻¹) therefore represents an NPP-based calculation included solely to illustrate the magnitude of overestimation when these methodological limitations are not considered (Text S11; Figs. S18–S19). Third, the machine-learning framework of **Chen et al. (2024)** represents an important methodological advance but differs from our approach in two key respects. Their method estimates Hg uptake flux by dividing foliar Hg pools by leaf lifespan, but assumes uniform leaf lifespan values for evergreen species (3–6 years), which does not capture the strong spatial variability in leaf longevity driven by climate and nutrient availability. In addition, leaf biomass is derived indirectly from leaf area index (LAI) and specific leaf area (SLA) (leaf biomass = LAI/SLA), a conversion that requires assumptions about canopy structure and vertical gradients in SLA. In contrast, our

framework harmonizes multiple foliar biomass datasets to produce observation-constrained estimates of leaf biomass, thereby avoiding uncertainties associated with indirect LAI–SLA conversions.

Revision in manuscript (Section 2.4, Lines 271–316):

The manuscript has been revised to clarify the methodological differences among existing modeling approaches, including mechanistic atmospheric models, NPP-based statistical methods, and recent machine-learning frameworks.

Revision in Supplementary Information (Text S11):

Additional explanation has been added describing the methodological limitations of NPP-based approaches and the rationale for presenting the NPP-derived estimate only as a methodological comparison (Figs. S18–S19).

2) In a related comment, the authors mention an “artificial overestimation of tropical uptake” from past studies. However, it is not clear to me in the paper or the SI where this overestimation in tropical uptake derives from and why it is “artificial” and not just resulting from decisions in the methodologies. This allegation needs to be further expanded, since it may be the source of the discrepancies between the estimates in the current study and others in the literature.

Response:

We thank the reviewer for raising this point. By “artificial overestimation of tropical uptake” we refer to methodological assumptions in some statistical approaches that systematically inflate uptake estimates in tropical regions, rather than reflecting true observational uncertainty.

A major source of this bias arises from the estimation of leaf biomass in NPP-based approaches. For example, Zhou and Obrist (2021) estimated foliar Hg uptake by multiplying foliar Hg concentrations by leaf net primary production (NPP), where leaf NPP was derived from total aboveground NPP using fixed allocation coefficients. This approach introduces two important biases in tropical forests. First, fixed allocation coefficients do not capture the higher and more variable leaf allocation typical of tropical forests (25–55%) compared with temperate and boreal systems (15–40%). Second, the foliar Hg concentrations used in these calculations represent multi-year cumulative accumulation in mature leaves. In tropical evergreen forests, where leaf lifespans commonly range from 1–3 years, multiplying these cumulative concentrations by annual NPP effectively counts multiple years of Hg accumulation within a single year.

A direct comparison of leaf biomass estimates illustrates the magnitude of this bias. Our harmonized leaf biomass dataset, which integrates multiple observational products (Text S3), yields tropical forest leaf biomass values of 6–10 t ha⁻¹. In contrast,

the NPP-based approach used by Zhou and Obrist (2021) implies leaf biomass values approximately 3–8 t ha⁻¹ higher, corresponding to a 30–80% overestimate of the leaf pool available for Hg accumulation (Fig. S18). This bias, combined with the use of cumulative foliar Hg concentrations, explains why Zhou and Obrist (2021) reported a higher global foliar Hg uptake estimate (1466 ± 490 Mg yr⁻¹) compared with our estimate (1119 ± 334 Mg yr⁻¹), with most of the discrepancy occurring in tropical regions.

Revision in manuscript (Section 2.4, Lines 287–303) and Supplementary Information (Text S11):

The manuscript and Supplementary Information have been revised to clarify that the apparent overestimation of tropical Hg uptake in some statistical approaches is methodological, arising from biased leaf biomass estimation using fixed allocation coefficients and the combination of cumulative foliar Hg concentrations with annual leaf NPP in tropical evergreen forests. A quantitative comparison of leaf biomass estimates is also provided (Fig. S18).

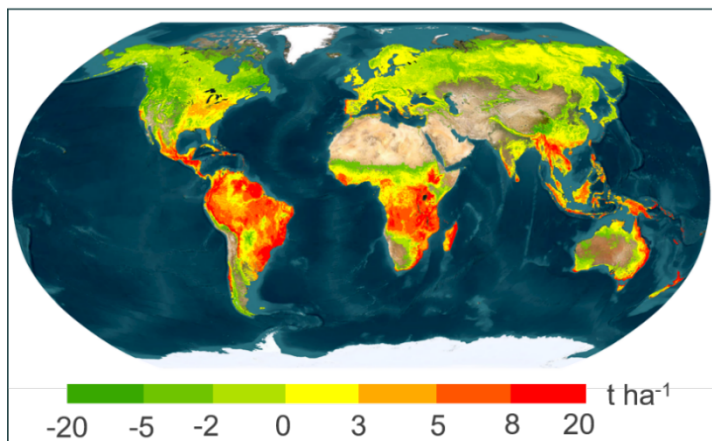


Figure S18. Difference between global foliar NPP (2010–2024) and modelled foliar biomass in this study. Foliar NPP was derived from aboveground NPP using leaf allocation fractions for four forest types (evergreen needleleaf: 0.46; deciduous needleleaf: 0.56; evergreen broadleaf: 0.58; and deciduous broadleaf: 0.64; Cleveland et al., 2013¹⁶) and weighted by forest-type-specific tree density³⁹. The resulting foliar NPP was then converted to foliar biomass using a fixed carbon fraction of 0.5.

3) One of the novelties of this study is the incorporation of leaf age into the calculation of forest uptake. This relies on the linear assumption of the foliar Hg accumulation rate over time. However, in Fig. 1c, it appears that none of this joint leaf age & concentration data is available from the Amazon or Africa, and for broadleaf trees all data is for leaves under 1 year of age. How do we know that accumulation effect doesn't saturate at older ages for broadleaf trees?

Response:

We acknowledge the reviewer's concern that direct observations linking leaf age to foliar Hg concentrations are currently unavailable for Amazonian and African tropical

forests, and that globally most available measurements for broadleaf species are limited to leaves younger than one year (Fig. 1c). While this represents an important data gap, we conducted several indirect validations to assess the robustness of the assumed linear accumulation of foliar Hg with leaf age.

First, for subtropical and temperate broadleaf species where age-resolved observations are available (up to 6 months for deciduous species and up to 1–2 years for some evergreen broadleaf species), foliar Hg concentrations increase approximately linearly with leaf age ($R^2 = 0.89\text{--}0.97$ across sites; Figs. 1c–d), with no indication of saturation within the observed age ranges. This pattern is consistent with the understanding that Hg^0 uptake occurs primarily through passive diffusion associated with stomatal gas exchange, which allows accumulation to continue as long as stomata remain active and atmospheric Hg^0 is available.

Second, we compared modeled foliar Hg uptake for evergreen broadleaf forests with independent litterfall Hg deposition measurements from subtropical and tropical regions. Across 18 sites with paired observations, modeled foliar uptake agrees closely with measured litterfall Hg deposition (slope = 0.96, $R^2 = 0.96$; Fig. S21). This agreement provides strong indirect support for the assumption that foliar Hg accumulation remains approximately linear over the lifespan of evergreen leaves in tropical forests.

Nevertheless, direct measurements of age-resolved foliar Hg concentrations in tropical forests remain a priority for future research. Such observations would allow explicit validation of the accumulation assumption and further improve model constraints.

Revision in manuscript (Section 2.4, Lines 339–361):

A paragraph has been added discussing the indirect validation of the linear foliar Hg accumulation assumption using age-resolved observations and litterfall Hg deposition comparisons.

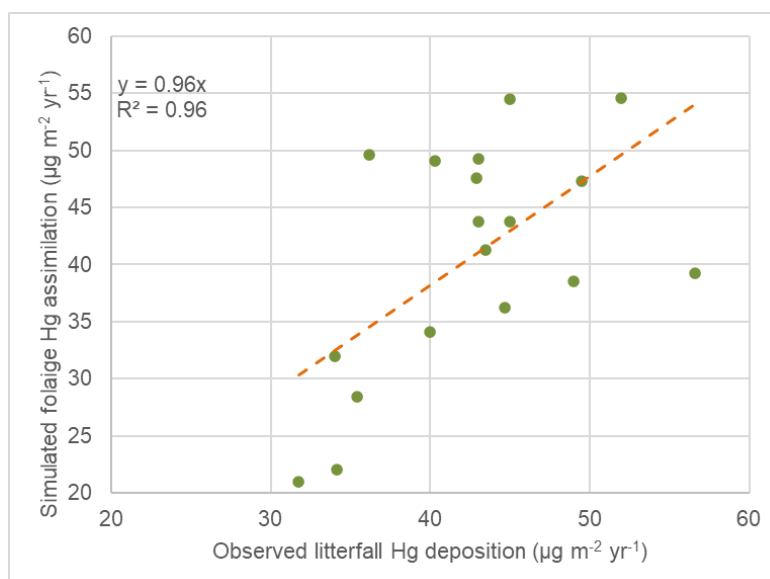


Figure S21. Comparison of modeled foliar Hg⁰ uptake in tropical evergreen broadleaf forests (Africa and the Amazon) with observed litterfall Hg deposition. The strong agreement indicates that the model captures Hg uptake in tropical forests despite limited observational coverage.

4) The methods section in the main manuscript is too brief to properly understand the approach; the salient information should be transferred from the SI into the main manuscript. For example, Section S3 in the SI is repetitive with what is already in the methods section but it includes additional information that should be incorporated into the main manuscript.

Response:

We thank the reviewer for this suggestion. The Methods section in the main manuscript has been revised accordingly, and the key information previously provided in Section S3 of the Supplementary Information has now been incorporated into the main text.

Revision in manuscript (Methods, Lines 477–478):

Key methodological information previously described in Section S3 of the Supplementary Information has been incorporated into the Methods section of the main manuscript.

5) Fig S25-36 - It says the model performance was evaluated on a test set, but in the methods it is stated that training is done through cross-validation. Was an independent evaluation dataset held separately as a test set after training? What percentage of the dataset was reserved and how was it selected? This should be clarified in the methods.

Response:

We thank the reviewer for requesting clarification of the model validation procedure. The Methods and Supplementary Information have been revised to explicitly describe the cross-validation approach used to evaluate model performance.

Revision in manuscript (Supplementary Information, Text S9, Lines 312–324):

The cross-validation procedure is now described as follows:

“Model performance was evaluated using 5-fold cross-validation rather than a single independent test set. The full dataset was randomly partitioned into five equal-sized subsets. For each fold, 80% of the data (four subsets) was used for model training and the remaining 20% (one subset) was used as the test set. This process was repeated five times, with each subset serving as the test set once. Performance metrics shown in Figs. S30–S50 represent aggregated predictions from all folds, ensuring that each observation is evaluated using an independent test prediction. Model performance was assessed using the coefficient of determination (R^2) and root mean square error (RMSE), together with residual diagnostics to examine error distribution and variance homogeneity.”

6) Figure 4 - This figure could be a bit confusing as the budget does not balance, since it only represents elemental Hg^0 in the Hg cycle. There is a deficit of 2000 Mg yr^{-1} , presumably coming from oxidation of Hg^0 to HgII . I also find confusing the manipulation of the Outridge et al. (2018) oceanic fluxes to produce this Hg^0 number, based on multiplying fluxes by a factor of 0.58 due to the isotopic source-tracing measurements in Jiskra et al. (2021). This assumption is inaccurate, since Outridge et al. (2018) refers to the net deposition flux (due to HgII dry and wet deposition to the ocean) and the net evasion flux (due only to Hg^0), with Jiskra et al. (2021) referring to 58% being the total of gross (i.e., one-way) deposition to the ocean. Thus, the budget in this figure leads to incorrect numbers resulting from mixing assumptions for net vs. gross fluxes and Hg^0 vs. HgII fluxes from different studies. I think it is misleading to modify the fluxes in this way, as these oceanic fluxes are not the focus of this work but presenting them in this Hg^0 budget could lead to these new estimates being compiled incorrectly by future studies.

Response:

We thank the reviewer for this critical and constructive comment. We agree that the original presentation of Fig. 4 could be misinterpreted as a complete Hg mass balance. The apparent deficit of $\sim 2000 \text{ Mg yr}^{-1}$ arose because the diagram focused exclusively on Hg^0 fluxes and did not include other important atmospheric Hg deposition pathways such as throughfall deposition, direct wet and dry deposition of Hg^{2+} to land and water surfaces, and atmospheric Hg^{2+} deposition to oceans.

We also agree that our previous modification of the oceanic flux estimates from Outridge et al. (2018) using the isotopic constraints from Jiskra et al. (2021) was not appropriate. Outridge et al. (2018) report net ocean–atmosphere Hg exchange, whereas Jiskra et al. (2021) quantify the fraction of gross atmospheric Hg deposition to oceans derived from anthropogenic sources. Because ocean Hg cycling is not the focus of this study, we have removed all oceanic flux estimates from Fig. 4 to avoid potential misinterpretation.

Revision in manuscript (Fig. 4 and caption, Lines 637–648):

Figure 4 has been revised to present only the terrestrial Hg^0 budget components directly relevant to this study, including:

- (i) anthropogenic Hg^0 emissions (updated to the 2000–2020 mean from Muntean et al., 2024),
- (ii) our revised estimate of global forest foliar Hg^0 uptake ($1188 \pm 438 \text{ Mg yr}^{-1}$), and
- (iii) the latitudinal comparison between Hg emissions and forest foliar uptake across three latitude bands (20°S – 10°N , 10°N – 40°N , and 40°N – 70°N).

The revised figure now clearly indicates that it represents a partial Hg^0 budget focused on the terrestrial vegetation sink, rather than a complete global Hg mass balance.

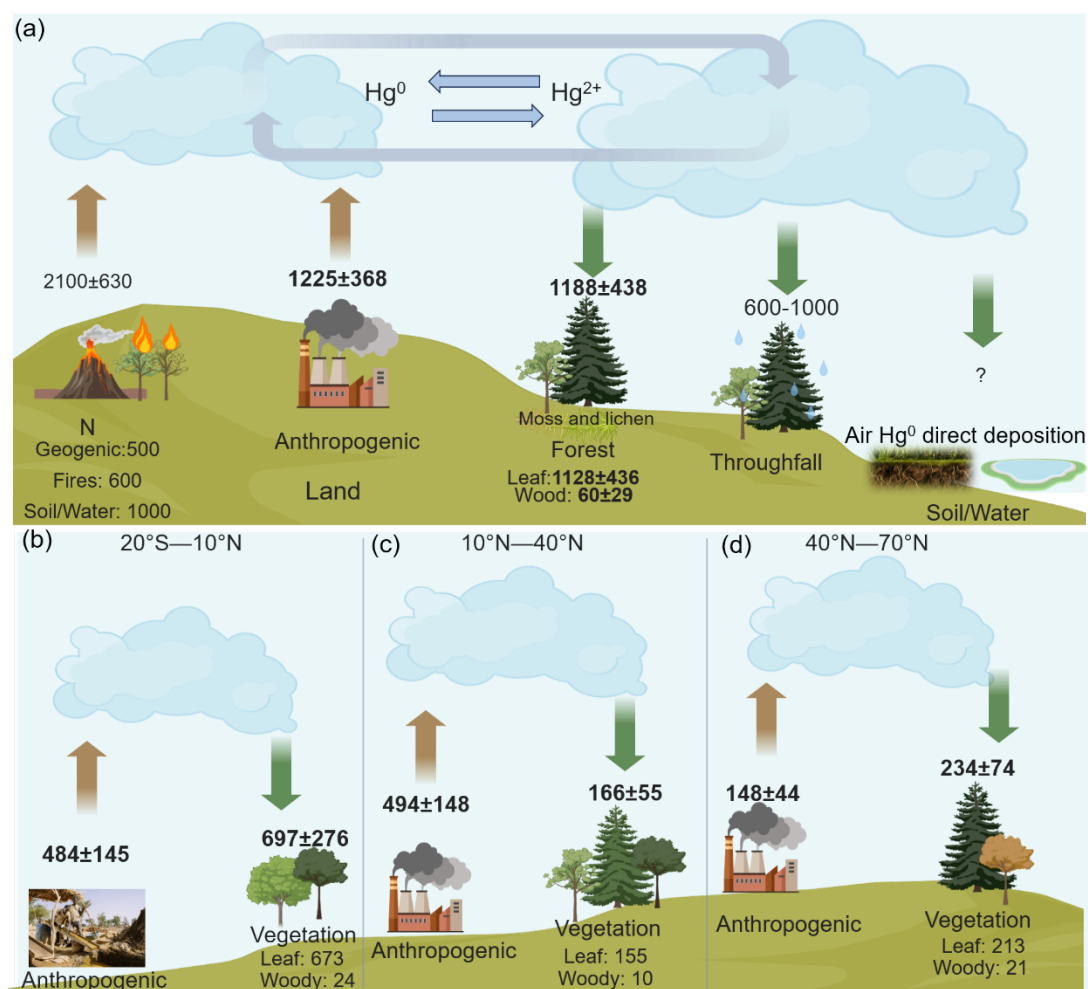


Figure 4. Global and latitudinal atmospheric Hg⁰ budgets (Mg yr⁻¹). (a) Global terrestrial Hg⁰ budget, modified from Outridge et al., 2018⁶², incorporating updated foliar Hg⁰ assimilation from this study and 2000–2020 anthropogenic Hg⁰ emissions⁴⁸. Atmospheric Hg⁰ deposition via throughfall are from Yuan et al. 2020⁵⁵. (b–d) Latitudinal Hg⁰ budgets showing mean anthropogenic Hg⁰ emissions over 2000–2022 and foliar Hg⁰ assimilation across (b) 20°S–10°N, (c) 10°N–40°N and (d) 40°N–70°N. Brown arrows indicate anthropogenic emissions, green arrows foliar assimilation, and silver arrows oceanic cycling. Uncertainties for Outridge et al. (2018)⁶² values were estimated as 30% of the mean. Abbreviation: N, non-current anthropogenic atmospheric Hg⁰ sources, including geogenic emissions, biomass burning and re-emissions from soils and waters, which partly recycle legacy anthropogenic Hg⁰. Note that this figure presents only Hg⁰ fluxes and does not represent a complete global Hg mass balance, which would require inclusion of HgII deposition and other flux pathways.

Other specific comments:

L63 - Jiskra et al. (2018) did a back-of-the-envelope calculation for the period 1990–2010, not the last two decades. A more recent study (Feinberg et al.,

doi:10.1073/pnas.2401950121, 2024) that investigated the trend over 2005–2020 using the GEOS-Chem model suggested that foliar uptake is not a major driver of recent Hg⁰ trends.

Response:

We thank the reviewer for pointing this out. Recent studies indicate that foliar uptake is unlikely to have been the dominant driver of the observed decline in atmospheric Hg⁰ concentrations during 2005–2020. To avoid potential misinterpretation, we have removed the statement suggesting that foliar uptake was responsible for the long-term decline in global atmospheric Hg⁰ levels.

Revision in manuscript (Introduction, Lines 61–62):

The sentence *“and has been identified as the driver behind the sustained decline in global atmospheric Hg⁰ levels between 1990 and 2010”* has been removed from the revised manuscript.

L82 - the GEOS-Chem parametrisation does not depend only on Hg⁰ concentration and leaf area index, there are other meteorological and land surface parameters included in the parametrisation.

Response:

We thank the reviewer for this helpful suggestion. The relevant sentence has been revised to clarify how current mechanistic models represent vegetation Hg uptake.

Revision in manuscript (Introduction, Lines 85–88):

The text now reads:

“Current mechanistic models such as GEOS-Chem incorporate multiple drivers of vegetation Hg uptake, including atmospheric Hg⁰ concentration, leaf area index, meteorological parameters (e.g., wind speed, temperature, humidity), and land surface characteristics (e.g., land use type and vegetation cover)^{24,27,29}. However, these models rely primarily on empirical parameterizations that do not explicitly represent key plant functional traits, particularly leaf age and stomatal conductance.”

Fig 2 - With the Structural Equation Modelling presented in Figure 2, it doesn't make sense to me that in plot a) climate has a negative relationship with anthropogenic activities, but in plot b) it has a positive relationship. Why should this change if these are independent from the broadleaf vs. coniferous distinction between plot a and b?

Response:

We thank the reviewer for identifying this inconsistency. Figure 2 has been revised to correct the relationships shown in the structural equation model so that they accurately reflect the SEM results.

L123- this must be a typo that deciduous broadleaves and evergreen needles have the exact same mean and standard deviation concentration ($31.1 \pm 18.8 \text{ ng g}^{-1}$)

Response:

We thank the reviewer for identifying this typographical error. The value has been corrected in the revised manuscript.

L243 - judging from Fig. S13, the order of the numbers cited in the brackets should be switched. 30.4 refers to China and 44.4 refers to Brazil. For clarity, it would be good to specify this specifically in the brackets, since otherwise it is not apparent that no measurements are available from central Africa

Response:

We thank the reviewer for pointing this out. The order of the values has been corrected and the locations (China and Brazil) are now explicitly indicated in the text to improve clarity.

-L275 - it should be emphasised somewhere that especially measurements in central Africa and tropical Southeast Asia are lacking in the database.

Response:

We thank the reviewer for this helpful suggestion. The manuscript has been revised to more clearly emphasize that measurements from central Africa and tropical Southeast Asia remain underrepresented in the current database, highlighting an important data gap in regions with potentially high Hg uptake.

-L280 - should be “inter-species” instead of “inter-specific”

Response:

We thank the reviewer for identifying this wording issue. The term has been corrected accordingly in the revised manuscript.

-L292, 293 - it's unclear where the emissions estimate comes from or what this refers to (is it total Hg emissions? Only primary anthropogenic? Terrestrial re-emissions?). Also, from what I can find in the Gerson et al. 2022 paper, there are no direct field measurements of emissions in that study, I think the 45 t yr^{-1} estimate is based on a bottom-up inventory estimate (cited refs 54 and 55 in Gerson22 study). It should be specified here where these numbers come from.

Response:

We thank the reviewer for requesting clarification of the emission estimates. Anthropogenic Hg^0 emissions were obtained from the EDGAR v8.1 emission inventory (2000–2020), which provides spatially resolved estimates of local anthropogenic Hg emissions (https://edgar.jrc.ec.europa.eu/dataset_tox81). The estimate of 45 t yr^{-1} for the Peruvian Amazon was derived from Gerson et al. (2022), which reports that

approximately 180 t yr^{-1} of Hg are used in artisanal and small-scale gold mining (ASGM) in the region, with roughly 25% emitted to the atmosphere, yielding an estimated atmospheric emission of $\sim 45 \text{ t yr}^{-1}$ (180×0.25).

Revision in manuscript (Section 3, Lines 372–375):

The data sources and calculation of regional Hg emissions have been clarified.

-L294 - what is the basis of the regional emissions number being $181 \pm 54 \text{ t yr}^{-1}$? What year is this based on? What inventory is this based on?

Response:

We thank the reviewer for this helpful question. The value $181 \pm 54 \text{ t yr}^{-1}$ represents the mean regional anthropogenic Hg^0 emissions in the Amazon averaged over 2000–2020, derived from the EDGAR v8.1 emission inventory. This information has been clarified in the revised manuscript (line 375-378).

-L320 - I can't find this 484 Mg yr^{-1} number in either the cited Outridge or Gerson papers. Where does it come from? Please specify the inventory used for calculating these numbers.

Response:

We thank the reviewer for this helpful question. The value 484 t yr^{-1} represents the mean anthropogenic Hg^0 emissions in tropical forest regions (20°S – 10°N) averaged over 2000–2020, derived from the EDGAR v8.1 emission inventory. This information has now been clarified in the revised manuscript (line 423-426).

-The references for Gerson et al. (2022) are repeated (50 and 52) in the reference list

Response:

We thank the reviewer for pointing this out. The duplicate reference for Gerson et al. (2022) has been removed from the reference list.

Table 1 - Yuan et al. 2024 didn't use GEOS-Chem, they used the Community Land Model–Hg (CLM5-Hg) within the Community Earth System Model (CESM).

Response:

We thank the reviewer for pointing this out. The description has been corrected to indicate that Yuan et al. (2024) used the Community Land Model–Hg (CLM5-Hg) within the Community Earth System Model (CESM) rather than GEOS-Chem.

-Section S3 - The clarity of the equations needs to be improved: 1) the authors denote i as age, but between t_1 and t_2 the age of the leaf will change. 2) Another issue in Section S3 is the “mass conservation principle” on Line 99: it is stated that difference between the foliar mass at time 1 and the foliar mass at time 2 is litterfall, but wouldn't

another change in the mass be due to photosynthesis? 3) Eq. 9 has non-Latin characters.

Response:

We thank the reviewer for these helpful comments.

(1) The notation has been revised to clearly distinguish leaf age at different times, using separate symbols to represent leaf age at t_1 and t_2 .

(2) The equation describes the net change in foliar biomass between t_1 and t_2 . Processes such as photosynthetic carbon assimilation are therefore implicitly included in the biomass term. This point has been clarified in the revised text.

(3) The formatting issue in Eq. 9 has been corrected and the non-Latin characters have been replaced with standard notation.

-The SI could be reorganised for easier navigation. Figures are cited out of order in the text (e.g., Fig. S47 mentioned before Fig. S30 for the first time, many of these examples). The numbers given on the title page do not correspond to the actual numbers of Figures (7 vs. 47).

Response:

We thank the reviewer for this helpful suggestion. The Supplementary Information has been reorganized to improve clarity and navigation. Figure citations have been corrected to follow a consistent numerical order and the figure numbering and counts on the title page have been updated to correspond to the actual number of figures in the SI.

-Fig S2 - I wonder if the air Hg concentration vs. foliage Hg concentration would be compared for individual sites rather than lumped over continents whether a relationship between these variables would be more apparent. Sites in Tibet will be quite different in air Hg concentration compared to Eastern China, and sites in industrialised or contaminated areas of Europe will also show higher air concentrations than elsewhere. Also, please clarify whether the air Hg concentrations that were analysed come from measurements or a model simulation.

Response:

We thank the reviewer for this thoughtful suggestion. We agree that site-level comparisons provide a more rigorous test of the relationship between atmospheric Hg^0 and foliar Hg concentrations than continent-scale aggregations. To address this point, we analyzed paired observations of atmospheric Hg^0 concentrations and foliar Hg measured at the same sites.

The analysis reveals a threshold response: when atmospheric Hg^0 concentrations exceed 5 ng m^{-3} , they exert a significant influence on foliar Hg concentrations ($p < 0.01$), whereas below this level the relationship is not significant ($p > 0.05$) (Fig. S1). Because atmospheric Hg^0 concentrations in most forest regions globally are typically

below 5 ng m^{-3} , regional-scale variations in atmospheric Hg^0 do not significantly control foliar Hg concentrations (Fig. S2). Instead, the observed global patterns of foliar Hg distribution are primarily governed by vegetation functional traits and climatic controls. Only in the immediate vicinity of strong point sources (e.g., ASGM sites, chlor-alkali plants, or urban emission centers) does atmospheric Hg^0 concentration become the dominant driver.

Revision in manuscript (Section 2.1, Lines 116–123):

The analysis of paired site-level observations of atmospheric Hg^0 and foliar Hg has been added to clarify the threshold relationship and its implications for global foliar Hg patterns.

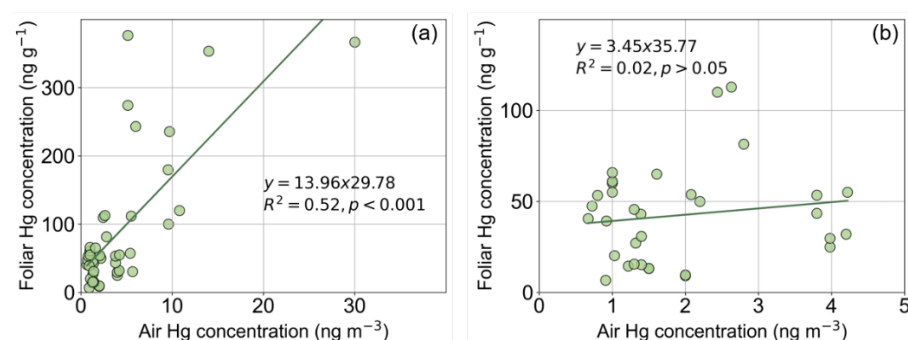


Figure S1. Relationship between atmospheric Hg concentration and foliar Hg concentration derived from co-located field measurements. (a) Relationship across the full atmospheric Hg concentration range. (b) Relationship restricted to background atmospheric Hg conditions (0–5 ng m⁻³). These results indicate that atmospheric Hg^0 constrains foliar Hg only above a threshold, while below it other controls dominate.

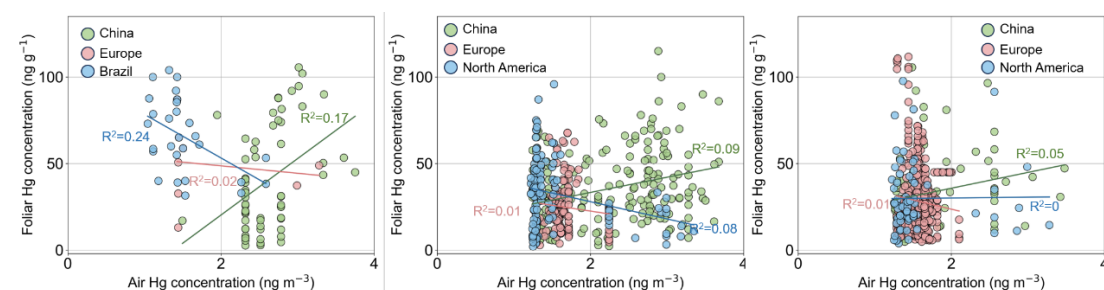


Figure S2. Relationships between foliar Hg concentrations and modeled atmospheric Hg^0 concentrations across different regions and forest types. Panel (a) shows evergreen broadleaf forests, panel (b) shows deciduous broadleaf forests and panel (c) shows evergreen needleleaf forests. These relationships show weak or absent correlations under background conditions, indicating limited regional control by atmospheric Hg^0 .

- The results in Fig S11 seem inconsistent to me with the approach used to calculate leaf age. The results show that the calculated foliar assimilation fluxes = 82% of litterfall flux (based on the slope of the fit). So that means that on average, litterfall has gained Hg from somewhere, e.g., post-depositional accumulation of atmospheric Hg. However,

in other parts of the paper (e.g. SI L167-168), it is assumed that litterfall represents the cumulative Hg level at the end of the leaf lifespan (not including post-lifespan accumulation in Hg?)

Response:

We thank the reviewer for this important observation regarding the relationship between modeled foliar Hg uptake and measured litterfall Hg fluxes. Following the reviewer's suggestion, we re-analyzed this relationship using a regression through the origin ($y = ax$), which imposes the physically appropriate zero-intercept condition.

With this revised approach, the slope of the relationship is 0.92 ($R^2 = 0.93$; Fig. S15), indicating that modeled foliar Hg uptake explains approximately 92% of the variance in measured litterfall Hg fluxes. The two estimates therefore agree within approximately 8% on average, providing strong support that our modeling framework accurately captures the magnitude of Hg transfer from forest canopies to soils through litterfall.

Revision in manuscript (Section 2.3, Lines 242–245):

The comparison between modeled foliar Hg uptake and litterfall Hg deposition has been revised using regression through the origin, and the updated slope and interpretation are now reported in the manuscript and Fig. S15.

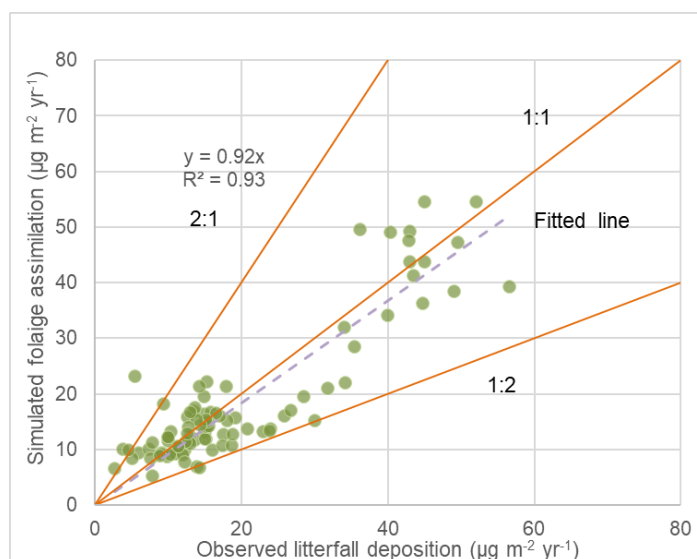


Figure S15. Comparison of modeled foliar Hg⁰ uptake with observed litterfall Hg deposition. The close agreement supports a near balance between canopy Hg uptake and litterfall export at annual timescales.

-Fig. S40 and S41 - It looks like samples with higher Hg concentrations are generally underestimated by the model, and the residuals increase with higher predicted values. Could this not affect the global estimate of Hg uptake? This needs to be further clarified since the foliar Hg concentration is a critical variable for this study

Response:

We thank the reviewer for this helpful comment. In response, we further optimized the model, which improved the overall predictive performance (R^2 increased from 0.60 to 0.63) and reduced the underestimation previously observed at higher foliar Hg concentrations. Some residual heteroscedasticity remains, which is expected because prediction errors tend to scale with concentration magnitude and because observations at the high end of the concentration range remain relatively limited.

A sensitivity analysis indicates that this residual variability does not materially affect the global estimate of forest foliar Hg^0 uptake, with differences of <5%. Nevertheless, we agree that additional observations from high-uptake regions—particularly tropical forests—would further improve model constraints and reduce uncertainty at the upper end of the concentration range.

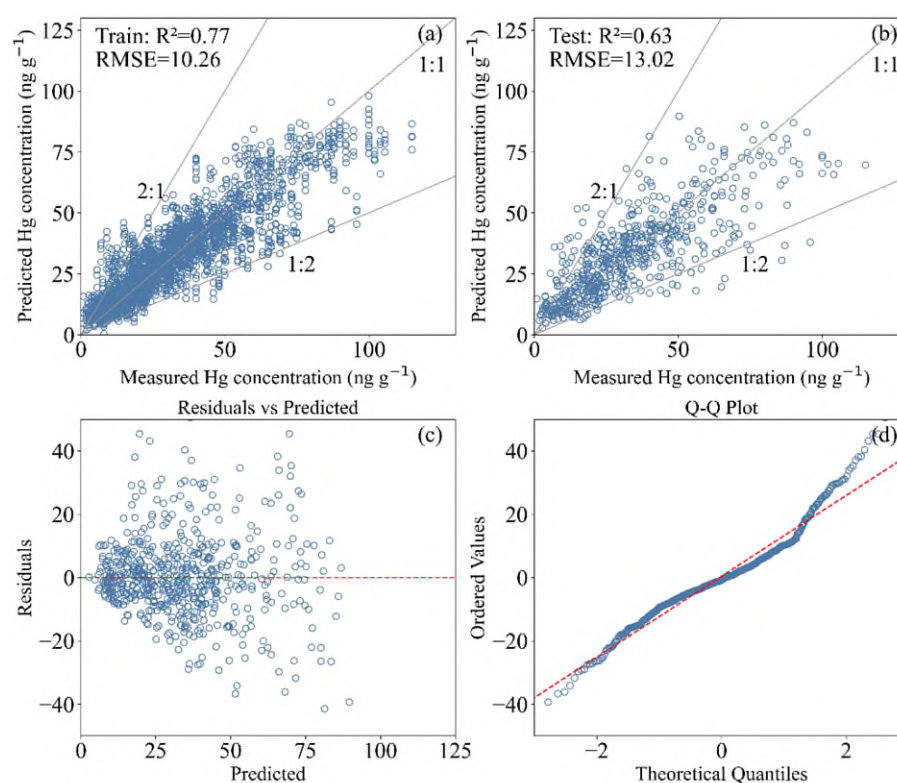


Figure S45. Model performance of foliar Hg concentrations for broadleaf forests. (a) Training dataset; (b) testing dataset; (c) scale–location plot; (d) Q–Q plot.

-SI Table 3 - What is the source of the gridded atmospheric Hg concentration data? Is it a model simulation or based on observations? If so, which model and what are the conditions of this run? This needs to be described somewhere as it is relevant for the conclusion that air Hg concentrations do not play a notable role in the spatial pattern of foliar uptake.

Response:

We thank the reviewer for requesting clarification of the atmospheric Hg concentration

dataset. The gridded atmospheric Hg⁰ concentrations used in this study were derived from the global mercury modeling framework described by Dastoor et al. (2025) (DOI: 10.5194/gmd-18-2747-2025). This dataset is based on simulations from multiple three-dimensional atmospheric Hg models (e.g., GEOS-Chem, GEM-MACH-Hg, and GLEMOS) coordinated within the Multi-Compartment Hg Modeling and Analysis Project (MCHgMAP). These models simulate global Hg cycling by representing atmospheric transport, chemical transformation, and deposition processes and are driven by harmonized emission inventories and meteorological reanalysis data.

Revision in Supplementary Information (Table S3):

A description of the atmospheric Hg concentration dataset and its modeling framework has been added to the Supplementary Information.

Reviewer #2 (Remarks on code availability):

I did not see any code uploaded.

Response:

Revised as the code has been uploaded.

Reviewer #3 (Remarks to the Author):

This is a sophisticated modeling study which simulates global forest Hg uptake on a global scale, with a particular focus on tropical forests. The authors state that unexpectedly, they find that (minimally industrialized) rainforests of South America and Africa exhibit the world's highest atmospheric Hg accumulation rates, and that these forests disproportionately sequester Hg released from industrialized regions. The study constrains global forest Hg uptake to $1119 \pm 329 \text{ Mg yr}^{-1}$, and the authors highlight a vulnerability of tropical forests to anthropogenic Hg inputs and climate-induced shifts in vegetation and terrestrial Hg reservoirs.

Response:

We thank the reviewer for the positive and encouraging assessment of our study. The constructive comments have been carefully considered and are addressed below.

The study represents, in my view, the highly sophisticated and advanced modeling approach of atmospheric Hg uptake by vegetation, which globally is a dominant atmospheric Hg sink and is subsequently transferred to ecosystems via litterfall leading to ecosystem exposure to this pollutant. They achieve this by finding, and implementing, that "leaf traits", particularly accumulation rates of Hg in foliage combined with foliar lifespan, govern foliar Hg concentrations and hence uptake. This is not a new finding per se, and has been documented in previous studies that show that leaf accumulation rates, leaf age and functional plant types determine foliar Hg uptake and lead to inherent differences in Hg levels among different plant functional types (i.e., broadleaf evergreens, coniferous, and deciduous plants). What is unique here, however, is that the authors are able to refine these relationships using global databases and implement these into a global model of forest leaf lifespan. I am not an expert in carbon/vegetation modeling and estimates of forest/foliar lifespan, but I encourage the authors to ensure their modeled lifespan estimates are compared/discussed with previous results and publications on this topic as it is fundamental for their new modeling approach of Hg uptake.

Response:

We thank the reviewer for this insightful comment. We agree that the credibility of the modeled leaf lifespan estimates is critical for the reliability of the Hg uptake framework. To address this point, we added a comparison between our modeled leaf lifespan and previously reported estimates in the Supplementary Information.

Revision in Supplementary Information (Text S3, Lines 108–119):

The following comparison has been added:

"Our modeled leaf lifespan values are broadly consistent with global plant trait syntheses, such as the worldwide leaf economics spectrum (Wright et al., 2004)². However, a remote-sensing-based study for tropical regions reported longer leaf

lifespans of 21–48 months⁸, compared with our modeled range of 12–21 months. Estimating leaf lifespan in tropical forests from satellite-derived leaf area index (LAI) is challenging because persistent cloud cover and the complex, multilayer canopy structure can obscure signals of leaf demography and introduce substantial uncertainty⁹. In addition, although our machine learning framework integrates extensive global trait datasets, the limited availability of direct ground-based leaf lifespan observations in tropical forests introduces additional uncertainty, as reflected in the model performance for these regions ($R^2 = 0.55$)."

The approach is transformational and allows for an unprecedented spatial and temporal method to simulate forest Hg update across global ecosystems. On a minor note, I noted that the authors in several places, including in the abstract, use the term physiological controls; in my view, the correct terminology is "trait-based framework (or functional/morphological plant traits), which the authors may want to use consistently throughout the manuscript as physiological data are not measured (only inferred).

Response:

We thank the reviewer for this helpful suggestion. We agree that the term *trait-based framework* more accurately reflects the approach used in this study. The manuscript has therefore been revised to replace the wording *physiological controls* with *trait-based framework (or plant functional traits*, where appropriate) to ensure consistent and precise terminology throughout the text, including in the abstract.

Revision in manuscript (Abstract and throughout the manuscript):

The terminology has been revised accordingly to emphasize that the framework is based on plant functional traits rather than directly measured physiological processes.

Previous studies have already highlighted the role of tropical forests as globally important atmospheric Hg sinks and hotspots for Hg deposition, so this is not new. This study, however, now reconciles results of previous studies based on a different scaling approaches (e.g., using NPP data) and correct large overestimation in some of these studies. The new global estimates are now quite consistent with ground-based litterfall deposition measurements/estimates - this is an important result. I also believe the authors have done a good job explaining uncertainties, variability, and potential error sources, which I appreciate. Because their modeling approach critically relies on accurate parameterization of leaf age/lifespans, however, I would appreciate a clearer description on how leaf age was measured/obtained/implemented from experimental databases into their model.

Response:

We thank the reviewer for this thoughtful and constructive feedback. We appreciate the reviewer's recognition of the importance of our reconciled global estimates and

their consistency with ground-based measurements. In response to this comment, we expanded the methodological description of the leaf lifespan dataset and modeling framework to improve transparency.

Revision in manuscript (Machine Learning, Lines 548–556):

The predicted leaf lifespan maps are used to parameterize the foliar Hg uptake model. Annual foliar Hg accumulation rates are calculated by dividing the Hg concentration in senesced foliage by the corresponding leaf lifespan, consistent with the approximately linear accumulation relationship observed between leaf age and foliar Hg concentration (Figs. 1c–1d).

Revision in manuscript (Supplementary Information Text S2):

Additional methodological details have been added describing the compilation of the leaf lifespan dataset, the predictor variables used in the modeling framework, and the model validation procedure. The dataset integrates global plant trait observations from the TRY Plant Trait Database and GLOPNET, supplemented by additional records retrieved from CNKI and Google Scholar, yielding 2,415 observational records after quality screening. Predictor variables include climatic factors (mean annual precipitation, temperature, solar radiation), spatial variables (longitude, latitude, elevation), and plant functional traits (specific leaf area, leaf nitrogen and phosphorus contents, and leaf area index). A machine learning model was developed to predict leaf lifespan at $0.1^\circ \times 0.1^\circ$ resolution and evaluated using 5-fold cross-validation, yielding R^2 values of 0.55 for evergreen broadleaf forests, 0.67 for deciduous broadleaf forests, and 0.64 for evergreen needleleaf forests.

The study would also benefit from some discussion of other deposition processes and deposition pathways, such as wet or Hg(II) deposition processes (e.g., in lines 288–291, the authors mention canopy interception and foliar fixation rates, yet it is unclear what processes dominates so some references to underlying uptake mechanism would be helpful).

Response:

We thank the reviewer for this valuable suggestion. To address this point, we compared our simulated foliar Hg⁰ uptake fluxes with published throughfall Hg deposition measurements.

Revision in manuscript (Section 3, Lines 382–388):

A comparison between modeled foliar Hg⁰ uptake and reported throughfall Hg deposition has been added (Fig. S22). The results show comparable magnitudes across climatic zones and similar latitudinal gradients, with the highest fluxes occurring in tropical forests. The text also clarifies that Hg(II) deposited via throughfall may be

more susceptible to lateral transport or rapid re-emission, whereas Hg transferred through litterfall is more effectively retained in soils through association with organic matter.

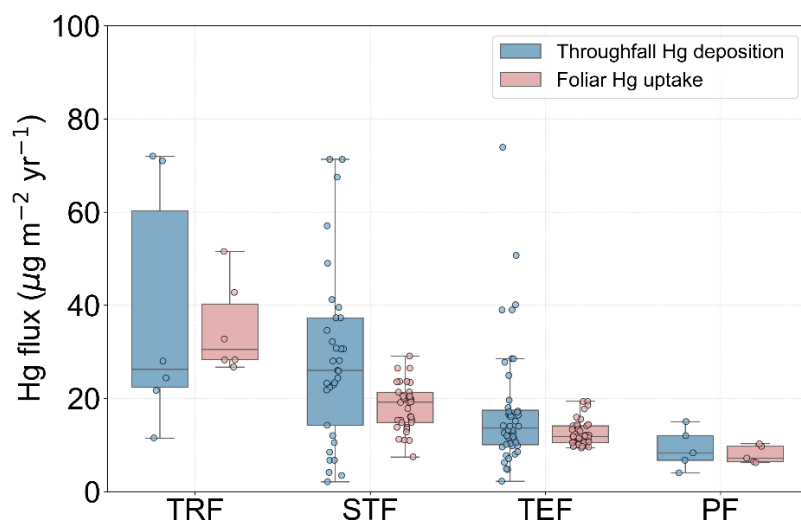


Figure S22. Measured throughfall Hg deposition and modelled foliar Hg⁰ uptake in this study across major latitudinal forest zones. TRF: Tropical forest; STF: Subtropical forest; TEF: Temperate forest; PF: Boreal (polar) forest. The consistent latitudinal gradients indicate that canopy uptake and throughfall reflect coherent patterns of atmospheric Hg inputs.

In lines 223-236, the authors discuss that annual uptake, litterfall, and net change in canopy (and woody pools) are near zero, so that globally and in tropical regions, these ecosystems are "stable, self-regulating interface that sustains the global balance between atmospheric Hg⁰ absorption and terrestrial deposition. Was this result modelled, measured, or simulated? This is unclear to me. I don't disagree with this notion in principle, but the authors should be careful to not underestimate (at least locally and regionally) impacts of interannual variability, climate variability, forest successional stages, or extreme climate/weather and disturbance events.

Response:

We thank the reviewer for raising this important point. The steady-state relationship between foliar Hg uptake and litterfall deposition was derived using a mass-balance framework assuming that, at the global scale, forest ecosystems operate near equilibrium over multi-year timescales. This interpretation is supported by the close agreement between modeled foliar Hg⁰ uptake ($1188 \pm 438 \text{ Mg yr}^{-1}$) and observed litterfall Hg deposition ($1180 \pm 710 \text{ Mg yr}^{-1}$), as well as their consistent spatial and latitudinal patterns (slope = 0.92, $R^2 = 0.93$; Fig. S15). We agree that this equilibrium assumption may not hold under all circumstances. Therefore, the manuscript has been revised to clarify that this quasi-equilibrium primarily applies to mature, undisturbed forests at the global scale, whereas climate variability, forest succession, and disturbances such as drought, fire, or deforestation may locally or regionally disrupt this balance.

Revision in manuscript (Section 2.3, Lines 240–250) and and Supplementary Information (Text S10, Lines 328–370):

The text has been revised to clarify the equilibrium assumption and to explicitly note that disturbances and environmental change may cause transient or long-term deviations from this balance.

My main concern with this study is with the interpretation and conclusion of results, particularly as they refer to fate of Hg and implication of this sink for tropical/global forests and global Hg cycling. The study exclusively focuses on the uptake and almost completely neglects discussing fate and emission/re-emission processes. The authors conclude that the tropics "disproportionately sequester Hg released from industrialized regions", "function as an unintended biogeochemical shield for the planet", that "absorbed Hg is largely stored in dynamic and climate-sensitive reservoirs, i.e. canopy, litter and organic-rich surface soils.", and that "legacy Hg has been redistributed via long-range transport and is now stored in remote tropical ecosystems". Yet, all these conclusions depend on understanding the fate and residence time of Hg in forests surface reservoirs and soils, after their original deposition via plant litterfall. The authors do not have data, nor do they discuss in detail, fate and turnover dynamics of Hg in forest soils, with the exception of mentioning one study from the the Peruvian Amazon which estimated that forest (floor?) emissions were in similar magnitudes as deposition. Do the authors suggest that this is the case across the tropics, and if so what is the basis is for this assumption?

Response:

We thank the reviewer for this valuable suggestion. In response, we expanded the discussion on the fate of mercury in tropical and global forests to clarify the processes governing Hg storage, re-emission, and long-term ecological implications.

Revision in manuscript (Section 3, Lines 382–397):

Additional discussion has been added describing Hg dynamics in tropical forest ecosystems. The revised text explains that high temperatures, humid conditions, and active microbial processes in tropical soils can promote Hg reduction and re-emission, yet sustained canopy inputs generally exceed these losses, resulting in net Hg accumulation in surface soils. The manuscript now also discusses the downward transport of Hg into deeper soil layers through precipitation-driven leaching and the stabilization of Hg in mineral soils. Furthermore, the text highlights that disturbances such as deforestation, fire, or prolonged drought may remobilize stored Hg, with observations showing soil Hg⁰ emissions increasing from ~6 $\mu\text{g m}^{-2} \text{yr}^{-1}$ before deforestation to ~230 $\mu\text{g m}^{-2} \text{yr}^{-1}$ after deforestation. The potential conversion of

inorganic Hg to methylmercury and its biomagnification through food webs is also discussed.

Revision in manuscript (Section 3, Lines 372–378):

We also clarified the interpretation of the Peruvian Amazon study, noting that the reported values largely represent areas strongly influenced by artisanal and small-scale gold mining (ASGM). In these locally contaminated regions, anthropogenic Hg emissions and forest Hg deposition can be of similar magnitude. However, this situation does not represent tropical forests more broadly. Across the Amazon Basin, forest uptake of atmospheric Hg^0 is estimated at $330 \pm 130 \text{ t yr}^{-1}$, exceeding regional anthropogenic emissions of $181 \pm 54 \text{ t yr}^{-1}$, indicating that forest deposition generally dominates the regional Hg budget outside heavily contaminated areas.

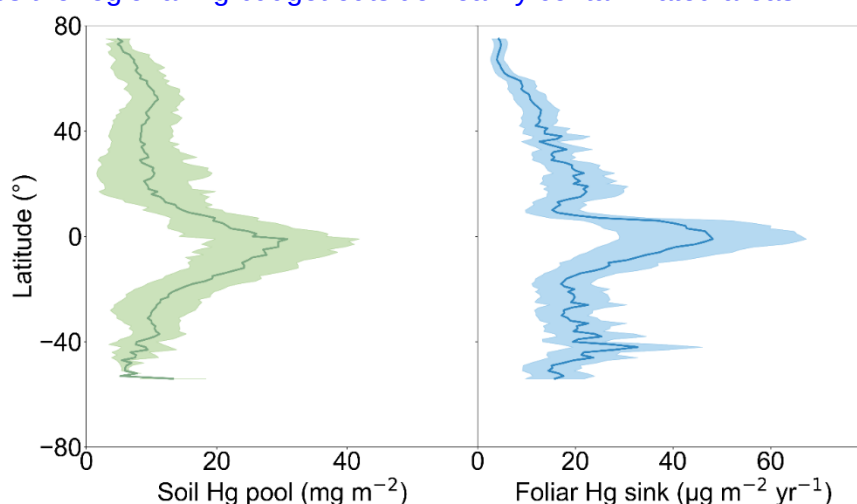


Figure S23. Latitudinal distribution of soil Hg pools⁴⁰ and modelled foliar Hg^0 uptake. The aligned latitudinal patterns suggest that foliar uptake followed by litterfall transfer governs surface soil Hg accumulation.

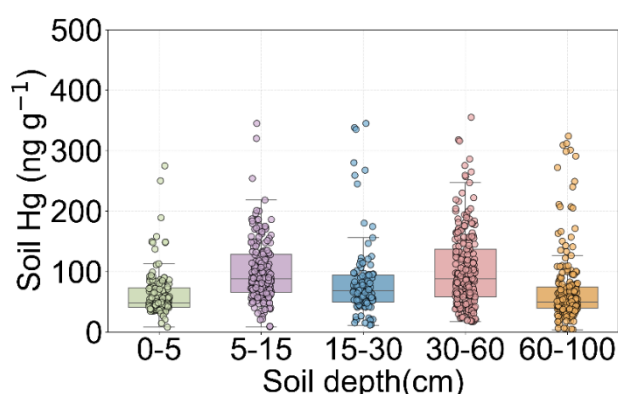


Figure S24. Soil Hg concentrations at different depth intervals. The vertical distribution indicates downward transport and retention of Hg from surface inputs into deeper soil layers.

Most of the discussion points related to equilibrium between deposition and emissions, and associated fate of Hg, is really confusing to me, and needs to be revised (e.g., lines 306-314. "near steady-state exchange"; forests "act as self-regulating buffers

rather than permanent sinks", "imply that terrestrial retention relies on intact canopy cover and sustained organic matter accumulation"). I cannot follow the underlying argumentation here. I recommend to consider an increasing data availability of soil Hg levels/pools, including from the tropics, in discussing fate and implication of high tropical atmospheric Hg deposition.

Response:

We thank the reviewer for this valuable suggestion. In response, we revised the manuscript to incorporate recent studies on forest soil Hg concentrations and pools, including observations from tropical regions. The revised text now discusses the fate of atmospheric Hg deposition in tropical forests in the context of soil Hg accumulation and soil–atmosphere exchange processes.

Revision in manuscript (Section 3, Lines 399-416; Supplementary Information Text S10):

The discussion now clarifies that although tropical forests receive some of the highest atmospheric Hg inputs globally, a substantial fraction of this Hg is retained in organic-rich surface soils, contributing to the large soil Hg pools observed in these ecosystems (e.g., Wang et al., 2019; Obrist et al., 2018). At the same time, continuous Hg⁰ re-emission from soils (e.g., ~63 $\mu\text{g m}^{-2} \text{yr}^{-1}$ in tropical forests) partially offsets the incoming flux, creating a dynamic quasi-equilibrium between deposition and emission at the soil–atmosphere interface. The revised text also emphasizes that intact forests can maintain this balance through sustained litterfall inputs and organic matter accumulation, whereas disturbances such as land-use change or climate extremes may disrupt this buffering capacity and alter the role of forests in the Hg cycle.

In summary, I find this an excellent and important study meritorious to be published in a highly visible journal such as Nature. The manuscript should be more specific about its limitations and conclusions, particularly as it refers to the importance of the findings for tropical forests health and long-term reservoirs/retention of global Hg pollution - this will critically depend on the stability of reservoir pools (surface organic layers and soils) which is not well considered/constrained in this study.

Response:

We thank the reviewer for the positive and encouraging evaluation of our work. In response to the reviewer's thoughtful suggestions, we revised the manuscript to clarify the limitations of our study and to refine the interpretation of tropical forests as long-term Hg reservoirs.

Revision in manuscript (Section 3, Lines 346–417):

The revised text now explicitly acknowledges several limitations and boundary conditions of our conclusions. First, while our model provides robust estimates of

global and regional foliar Hg⁰ uptake, direct observations of Hg retention, transformation, and re-emission in tropical soils remain limited, which introduces uncertainty in predicting long-term Hg storage. Second, the steady-state relationship inferred between foliar uptake and litterfall deposition primarily applies to mature, undisturbed forests under average climatic conditions, and may not hold during forest succession, extreme disturbances, or under future climate and land-use change. Third, the manuscript now emphasizes that although tropical forest soils accumulate substantial Hg inputs from litterfall, they should not be considered permanent sinks. Disturbances such as deforestation, fire, or hydrological perturbations may remobilize stored Hg, potentially converting these ecosystems from sinks to sources of Hg to the atmosphere and aquatic systems. The revised discussion highlights that the stability of these Hg reservoirs depends strongly on forest integrity, and that large-scale disturbances could influence the long-term effectiveness of Hg mitigation efforts such as the Minamata Convention.

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RESPONSE TO REVIEWERS' COMMENTS

Reviewer #2 (Remarks to the Author):

The authors have carefully addressed the reviewer comments, and I generally agree with the revisions that have been made. I have two remaining concerns:

1) During the revision, several of the linear regression analyses were adjusted to force the line of best fit to go through the origin (e.g., Fig S15 and S21). This is slightly misleading, as the R^2 for these fits become artificially inflated when the zero-intercept condition is used. The R^2 for these lines are no longer representative of goodness of fit, because they are calculated with the sum of squares using the origin instead of the mean of the data. I also don't see a physical justification here, as opposed to other cases where a zero-intercept condition would be employed, e.g., that distance = velocity * time. In the case of these two figures, observed litterfall deposition is compared to modelled foliage assimilation, with no clear reason for a zero intercept. Any modelled quantity could have a systematic offset from observations, and thus a non-zero intercept is highly probable and should be considered in regression models.

RE: We appreciate the reviewer's insightful comment. We have revised the regression analyses in Figs. S15 and S21 to ordinary least squares (OLS) regression with an intercept. We have also incorporated litterfall Hg flux data from three tropical montane evergreen forest sites in the eastern Congo Basin, Africa (Bouchet et al., 2026. High mercury stocks in eastern Congo basin soils and their substantial mobilization after deforestation). The corresponding figures and in-text descriptions have been updated accordingly.

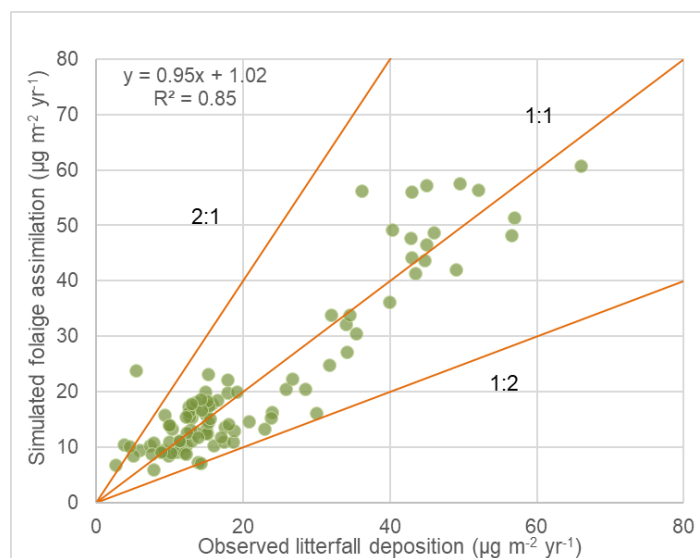


Figure S16. Comparison of modeled foliar Hg^0 uptake with observed litterfall Hg deposition. The close agreement supports a near balance between canopy Hg uptake and litterfall export at annual timescales.

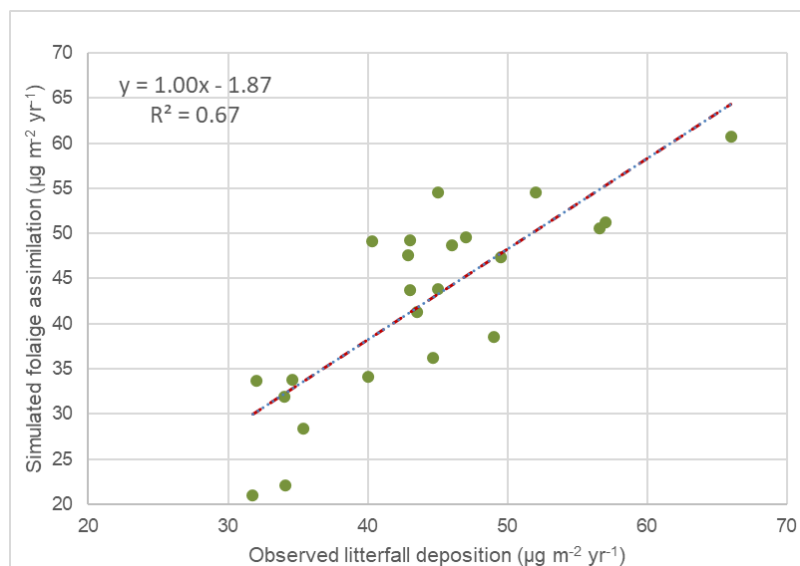


Figure S22. Comparison of modeled foliar Hg^0 uptake in tropical evergreen broadleaf forests (Africa and the Amazon) with observed litterfall Hg deposition. The strong agreement indicates that the model captures Hg uptake in tropical forests despite limited observational coverage.

2) In the new calculation of source-proximal areas, from Text S5 it looks like a dataset of all mining-impacted areas (Tang and Werner 2023) was used to extract areas over which to calculate the global emission-impacted flux. However, this dataset is a general dataset for mining and thus not specific to ASGM locations. I think a further clarification is needed of which emissions source impacts are treated in this calculation. Is it just meant to be ASGM, in which case a specific ASGM geographical distribution should be considered; or are other contaminated forest sites considered, in which case why is this general mining-impacted areas map an appropriate dataset to capture Hg contamination?

RE: We thank the reviewer for the request for clarification. We have thoroughly re-examined the leaf mercury (Hg) dataset collected from contaminated areas and the definition of source-proximal forests. The field-measured leaf Hg data from contaminated environments mainly fall into two categories: artisanal and small-scale gold mining (ASGM) areas, and mercury or gold mining areas. Given that ASGM is currently the significant anthropogenic source of global Hg emissions, we have revised our calculation to focus specifically on ASGM regions and mercury/gold mining areas—both of which are typical sources of Hg pollution—while other mining types are not considered at this stage due to insufficient observational data.

We no longer rely on the general mining-impacted area dataset (Tang and Werner, 2023) to extract all mining-influenced zones. Instead, we used the spatial

distributions specific to ASGM and mercury/gold mining to delineate source-proximal forest areas, with the datasets obtained from their respective source websites: the UNEP/MapX ASGM dataset (https://app.mapx.org/static.html?views=MX-DYYKK-FAWSZ-NXSIO&theme=color_light&zoomToViews=true) and the ICMM Global Mining Dataset (<https://www.icmm.com/en-gb/research/data/2025/global-mining-dataset>).

The leaf Hg uptake in these forests was estimated using two approaches:

1. A machine learning model built on all collected field observations, which yields a total leaf Hg uptake of $24.7 \pm 28.8 \text{ Mg yr}^{-1}$ for forests adjacent to ASGM and mercury/gold mining areas. Within this total, the uptake for ASGM-proximal forests is $19.6 \pm 23.2 \text{ Mg yr}^{-1}$ and for mercury/gold-mining-proximal forests is $5.1 \pm 5.6 \text{ Mg yr}^{-1}$.
2. Empirical estimates based on literature-reported leaf Hg concentrations combined with foliage biomass. Using the mean of the measured leaf Hg concentrations, the estimated uptake is $19.6 \pm 23.1 \text{ Mg yr}^{-1}$ for ASGM-adjacent forests and $14.9 \pm 18.5 \text{ Mg yr}^{-1}$ for mercury/gold-mining-adjacent forests. Using the median of the measured concentrations, the corresponding values are $10.4 \pm 9.6 \text{ Mg yr}^{-1}$ and $5.2 \pm 4.5 \text{ Mg yr}^{-1}$, respectively.

We note that the current estimate of leaf Hg uptake in source-proximal forests is still subject to high uncertainty. This uncertainty stems mainly from (1) the large inter-site variability in observed leaf Hg concentrations (ranging from 20 to 6654 ng g^{-1}), which makes extrapolation from limited observations challenging, and (2) the still-insufficient spatial coverage of measured leaf Hg data in ASGM and mercury/gold mining regions, particularly in areas with intense ASGM activity such as Africa and Indonesia, as well as in certain mercury/gold mining hotspots. Future field campaigns targeting leaf Hg in these source-proximal forests are needed to further constrain the estimate.

We have revised Text S5 and the corresponding supplementary figure (Fig. S10-S12) and lines 215-225 in the manuscript to explicitly specify the data sources, the refined definition of source areas, and the updated calculation results.

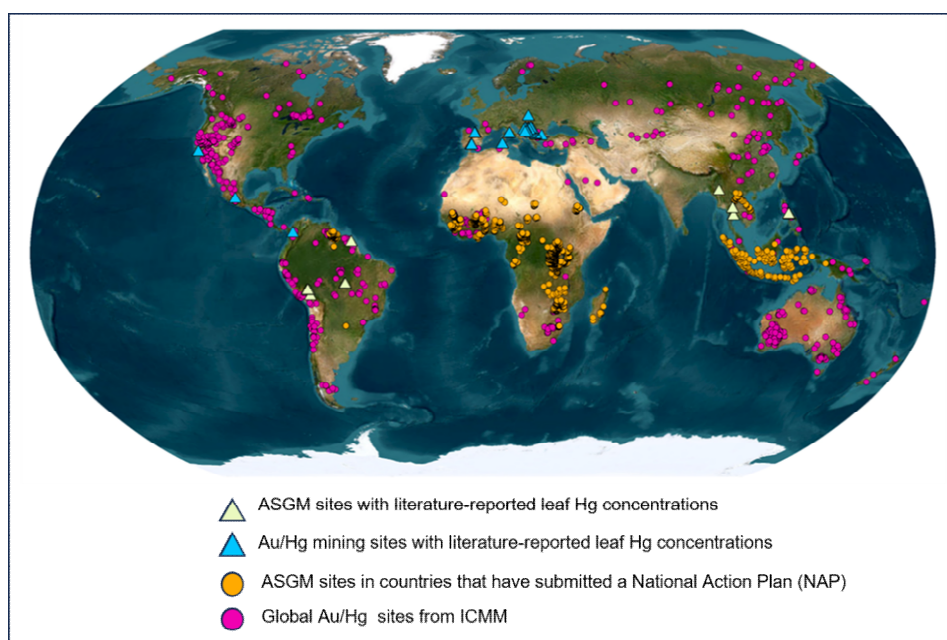


Fig. S10. Global distribution of ASGM sites, Au/Hg mines, and corresponding literature-reported leaf Hg concentration sites.

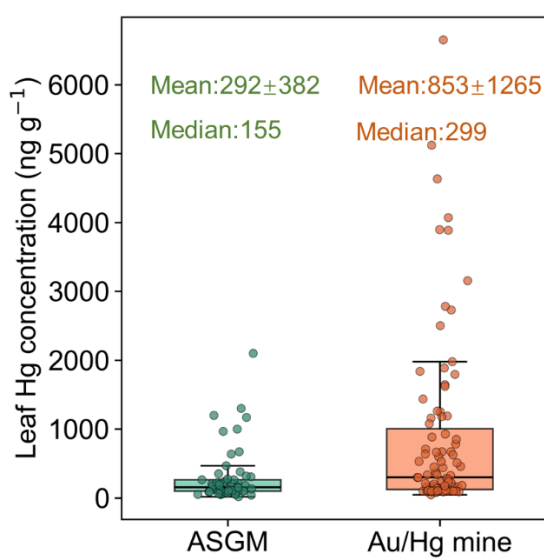


Fig. S11. Literature-reported leaf Hg concentrations at ASGM and Au/Hg mining sites.

Reviewer #2 (Remarks on code availability):

I have seen the code upload and it looks thorough, although I have not reviewed this in depth.

RE: Thanks.